ISSN: 2578-8760

Journal of Nanomedicine

Open Access | Research Article

Recyclability of Spray Coated Smooth Nanocellulose Films as a Potential Sustainable Alternative to Synthetic Food Packaging-A Perspective

Kirubanandan Shanmugam*

Saveetha School of Engineering, SIMATS, Thandalam, Chennai -602107, India.

***Corresponding Author(s): Kirubanandan Shanmugam**

Saveetha School of Engineering, SIMATS, Thandalam, Chennai-602107, India.

Email: kirubanandan.shanmugam@gmail.com

Received: Jan 06, 2024

Accepted: Feb 07, 2024

Published Online: Feb 14, 2024

Journal: Journal of Nanomedicine

Publisher: MedDocs Publishers LLC

Online edition: http://meddocsonline.org/

Keywords: Nanocellulose; Spray coating; Recycling; Water vapour permeability; Tensile Index.

Abstract

Synthetic packaging materials are neither reprocessable, renewable nor biodegradable barrier and also difficult to recycle into useful products. While cellulose-based packaging is widely known as a renewable, reprocessable, recyclable and biodegradable material it has limited air and water vapour barrier properties. Nanocellulose (NC) has the potential to become a renewable alternative to plastic packaging barrier layers but its recyclability has not been reported. This investigation is focused on the production of spray-coated Nanocellulose (NC) films and its recyclability via dispersion in water and vacuum filtration to form sheets again. Physical properties such as barrier performance and strength of NC films have been evaluated pre- and post- recycling. The recycled films retained 70% of tensile strength and significant barrier performance although the H_2O Vapour Permeability (WVP) approximately doubled, increasing to 9.83 x 10^{-11} $g.m^{-1}.s^{-1}$ Pa^{-1}, providing comparable values with for most synthetic packaging. SEM micrographs reveal no fibre agglomeration at the micro level and no damage to the fibres during recycling. The retained strength and barrier properties and facile reprocessability of the spray coated NC film promises a sustainable and recyclable alternative to conventional packaging, providing a sustainable platform for packaging industries.

Introduction

Food packaging materials used to protect the foods from water vapour and oxygen. These barrier materials protect the food stuff from the degradation. Some common food packaging materials include: Plastic: Polyethylene Terephthalate (PET), High-Density Polyethylene (HDPE), Polypropylene (PP), and Polystyrene (PS) are commonly used for food packaging. Plastic is lightweight, durable, and provides good moisture barrier properties. However, it can contribute to environmental pollution [1]. Glass: Glass containers are commonly used for food packaging, especially for beverages, sauces, and pickled products. Glass is inert, does not affect food quality, and can be easily recycled [2]. Metal: Aluminium and tin are commonly used for food cans, beverage cans, and aerosol cans. Metal packaging provides good protection against light, moisture, and oxygen. It is also recyclable [3]. Paper and cardboard: Paper and cardboard packaging is commonly used for dry food products like cereals, pasta, and snacks. These materials are renewable, biodegradable, and easily recyclable [4]. Flexible films: Flexible films made of various materials like laminated plastics, aluminium, and paper are used for packaging snacks, cookies, and other products.

Cite this article: Shanmugam K. Recyclability of Spray Coated Smooth Nanocellulose Films as a Potential Sustainable Alternative to Synthetic Food Packaging-A Perspective. J Nanomed. 2024; 7(1): 1066.

These films provide good barrier properties and extend the shelf life of the food [5]. Bio-based materials: Increasingly, bio-based materials like bioplastics, made from renewable resources like corn or sugarcane, are being used for food packaging. These materials offer similar properties to conventional plastics but are more environmentally friendly. Composite materials: Composite materials, such as tetra Pak, combine different layers of various materials like paper, aluminium, and plastic to create packaging that provides better protection against light, oxygen, and moisture. It's important to note that the choice of packaging material depends on the specific food product, its required shelf life, and the desired level of protection.

Biopolymer coatings refer to thin layers of biodegradable and renewable materials applied to the surface of food packaging materials. These coatings are used to improve the barrier properties, mechanical strength, and shelf life of the packaging materials, while also offering a more sustainable alternative to conventional synthetic coatings. Some common biopolymers used for food packaging coatings include: Polyhydroxyalkanoates (PHA): PHA coatings are derived from microbial fermentation of renewable feedstock and offer good barrier properties against gases, moisture, and aroma compounds. Polylactic Acid (PLA): PLA is a biodegradable polymer produced from renewable resources such as corn starch or sugarcane. It has good transparency and thermal stability, making it suitable for packaging applications. Chitosan: Chitosan is derived from chitin, a component found in the shells of crustaceans. It has antimicrobial properties, which can help extend the shelf life of packaged food products. Cellulose derivatives: Cellulose, obtained from plants or bacteria, can be chemically modified to form derivatives like cellulose acetate or methylcellulose. These derivatives improve the barrier properties of food packaging materials.

Benefits of biopolymer coatings for food packaging: Sustainability: Biopolymers are derived from renewable resources, reducing reliance on fossil fuels and contributing to a more sustainable packaging solution. Biodegradability: Biopolymer coatings are typically biodegradable, meaning they can be broken down by natural processes, reducing their environmental impact. Barrier properties: Biopolymer coatings can provide excellent barrier properties against moisture, gases, and other contaminants, helping to maintain the quality and freshness of food products. Consumer perception: Many consumers are increasingly concerned about the environmental impact of packaging materials. By using biopolymer coatings, food companies can cater to the demands of eco-conscious consumers. There are ongoing research and development efforts to improve the performance and functionality of biopolymer coatings for food packaging, including enhancing their mechanical properties, barrier performance, and process ability.

Nanocellulose is a promising material for packaging due to its unique properties and environmental benefits. It is derived from renewable sources like wood pulp and has a high strength-to-weight ratio, making it ideal for packaging applications. One of the main advantages of nanocellulose as packaging is its biodegradability. Unlike traditional plastic packaging, nanocellulose-based packaging materials can be broken down by microorganisms in the environment, reducing the environmental impact. Furthermore, nanocellulose films have barrier properties that can rival or even surpass those of conventional packaging materials. They can effectively prevent the migration of gases, such as oxygen and moisture, which helps in preserving the freshness and extending the shelf life of packaged products. Nanocellulose can also be modified to impart additional properties, like antimicrobial or antioxidant activity, making it suitable for food packaging applications. These characteristics can help in preventing microbial growth and maintaining food quality and safety. Additionally, nanocellulose films are transparent, which allows consumers to see the packaged product without compromising its protection. This feature is particularly important in the food industry, where visual inspection is often preferred. Moreover, nanocellulose-based packaging materials can be produced using sustainable manufacturing processes. The production of nanocellulose involves non-toxic and energy-efficient processes, which further contributes to its environmental friendliness. Overall, nanocellulose offers a sustainable and eco-friendly alternative to traditional packaging materials. Its unique properties, including biodegradability, barrier properties, and transparency, make it an attractive choice for various packaging applications.

Nanocellulose films are thin films made from nanocellulose materials. Nanocellulose is a nanomaterial derived from cellulose, which is the main component of plant cell walls. It is made up of tiny cellulose fibres that are several nanometres in width and several micrometres in length. The production of nanocellulose films involves the extraction of cellulose fibres from plant sources, such as wood or other biomass. These fibres are then broken down into smaller nanofibers using mechanical or chemical processes. The resulting nanofibers are then dispersed in a liquid medium, such as water, to form a suspension. The suspension is then cast onto a surface and dried, resulting in the formation of a thin film. The film retains the unique properties of nanocellulose, including its high strength, flexibility, transparency, and biodegradability. These properties make nanocellulose films highly versatile and suitable for a wide range of applications.

Nanocellulose films have diverse applications in various fields, including packaging, electronics, biomedical engineering, and environmental remediation. For example, they can be used as a sustainable alternative to petroleum-based films in food packaging to enhance barrier properties and preserve freshness. In electronics, nanocellulose films can be used as substrates for flexible displays and as energy storage devices. In biomedical engineering, nanocellulose films can be used as scaffolds for tissue regeneration, drug delivery systems, and wound dressings. They are also being explored for their antibacterial properties, which could enable their use in the development of antimicrobial coatings. Overall, nanocellulose films offer a promising solution for creating sustainable and multifunctional materials with a wide range of applications.

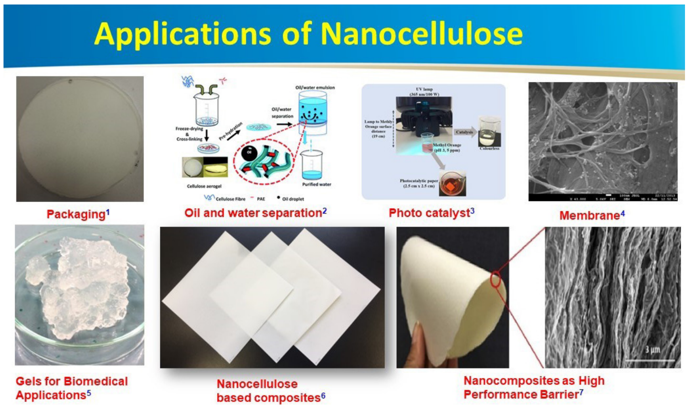


Figure 1: Application of nanocellulose and its films and composites.

Nanocellulose and its free standing films and composites was numerous in the field of material science and engg. Free standing nanocellulose film can be used as a potential barrier material and also good alternative for synthetic plastics. The nanocellulose film was used as membrane for the seperation of oil from oil water emulsion. The TiO_2 incorporated nanocellulose film was fabricated and used as photo catalyst for treatment of waste water. Nanocellulose gels used as blood test and diagostic medium in biomedical fields. Nancellulose -Inorganics composite such as Nanocellulose-Bismuth composite and Nanocellulose -Montmorinollite composite were used greener packaging materials.

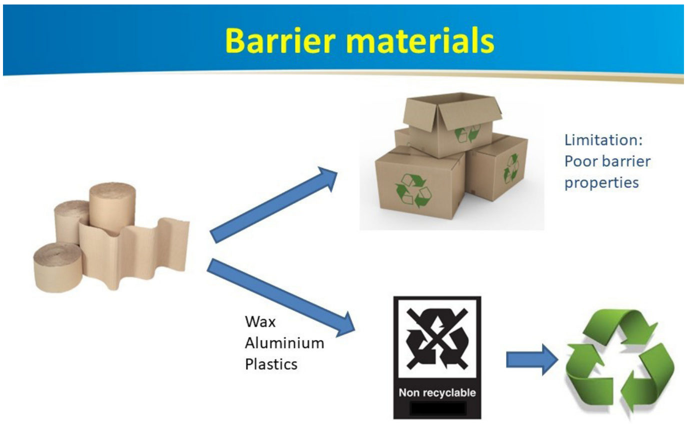


Figure 2: Necessary of coating of Paper and Paper board.

Barrier materials are substances that prevent or hinder the penetration or transfer of certain substances, such as liquids, gases, moisture, or light. They are commonly used in various applications to protect products, preserve freshness, maintain integrity, or enhance safety. There are different types of barrier materials available, each with its specific properties and applications. Some common examples include:Flexible Films: Films made of plastic or polymer materials that possess excellent barrier properties against gases, moisture, and odors. They are widely used in food packaging, pharmaceuticals, electronics, and agriculture.Metalized Films: Films that are coated with a thin layer of metal, usually aluminum, to provide enhanced barrier properties to light, gases, moisture, and UV radiation. They are commonly used in food and beverage packaging, such as coffee bags or snack packaging.Glass: Glass is an excellent barrier material against gases, liquids, and UV radiation. It is commonly used in the packaging of pharmaceuticals, perfumes, and high-end cosmetic products.Foil: Aluminum foil is a common barrier material that provides excellent protection against moisture, gases, and light. It is widely used in food packaging, particularly in the form of pouches, trays, and wraps.Paperboard: Paperboard materials with barrier coatings or laminations can provide moisture and grease resistance. They are commonly used in packaging for dry food products, such as cereals, snacks, and frozen food.Rubber and Elastomers: Rubber or elastomeric materials can act as barriers against gases, liquids, and moisture. They find applications in medical devices, automotive components, and industrial seals.Coatings: Specialized coatings can be applied to various substrates to provide barrier properties. These coatings can be moisture-resistant, solvent-resistant, or gas-resistant, depending on the specific application. Barrier materials play a crucial role in protecting products from external elements, extending their shelf life, minimizing spoilage, and ensuring safety. The selection of the appropriate barrier material depends on the specific requirements of the product and the desired barrier properties.

Figure 2 reveals the necessary of coating of paper and paper board. Normally, cellulose substrates such as paper and paper board has poor barrier peformance due to the presence of wide pores and surface pores in the substrates. These pores allow more air and water vapour across the substrates resulting poor barrier potential. To mitigate this problem, the paper substrated coated with synthetic platics,or extrusion with aluminum and wax coating for imporving their barrier peroformance. These coating and plastics are not recyclable and produce a theart to the environment. This is why, the coating of biopolymer on the paper substrates was attempted to promote their barrier potential. Recently, nanocelluose coating on the paper substrates was developed and these nanocellulose has potential in filling the surface pores of the paper and paper board and forms a barrier film on the paper substrates. The resulted coated substrates has excellent barrier performance against air and water vapour.

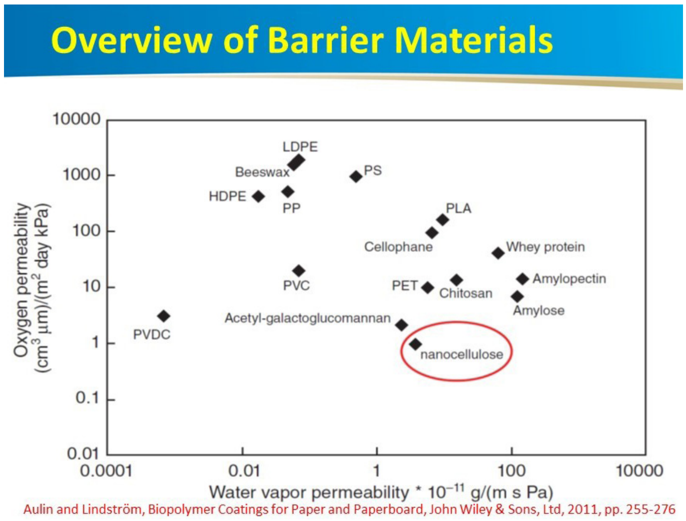


Figure 3: Barrier performance of Biopolymers and Synthetic Plastics.

The relationship between the biopolymer and synthetic polymers' water vapour and oxygen permeabilities is shown in Figure 3. The accompanying figure demonstrates that CNF has an excellent barrier against oxygen and outperformed other biopolymers, biodegradable materials, and synthetic plastics in barrier performance. However, compared to synthetic plastics, CNF/NC has a weak barrier against water vapour, a high water vapour permeability, and a low water vapour permeability when compared to other biopolymers.

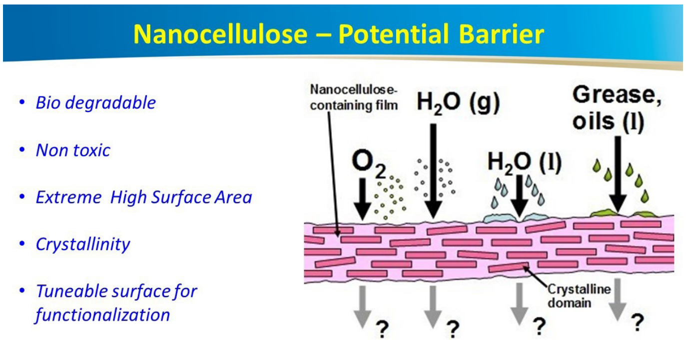


Figure 4: Barrier Potential of Nanocellulose.

The barrier potential of nanocellulose refers to its ability to act as a barrier against the flow of certain substances or to prevent the entry of external elements into a specific area. Nanocellulose, which is derived from cellulose fibers and has a nanoscale size, offers unique properties that make it an effective barrier material in various applications. One of the key factors that contribute to the barrier potential of nanocellulose is its high surface area. Due to its nanoscale dimensions, nanocellulose has an increased surface area compared to conventional cellulose materials. This increased surface area allows for a higher number of contact points and interactions, making it more difficult for substances to penetrate through the material. Furthermore, nanocellulose has a highly organized and dense structure, which further enhances its barrier properties. The nanofibers in nanocellulose can form a tight network, creating a physical barrier that restricts the movement of molecules and ions. This organized structure also provides mechanical strength to the material, making it resistant to deformation or damage. Nanocellulose can also be modified or functionalized to enhance its barrier potential. For example, coating nanocellulose with hydrophobic materials can improve its water vapor barrier properties, while incorporating antimicrobial agents can prevent the growth of microorganisms. These properties make nanocellulose suitable for various barrier applications, such as in packaging materials, protective coatings, filters, membranes, and biomedical devices. It provides an environmentally-friendly and renewable alternative to traditional barrier materials, offering improved performance and sustainability.

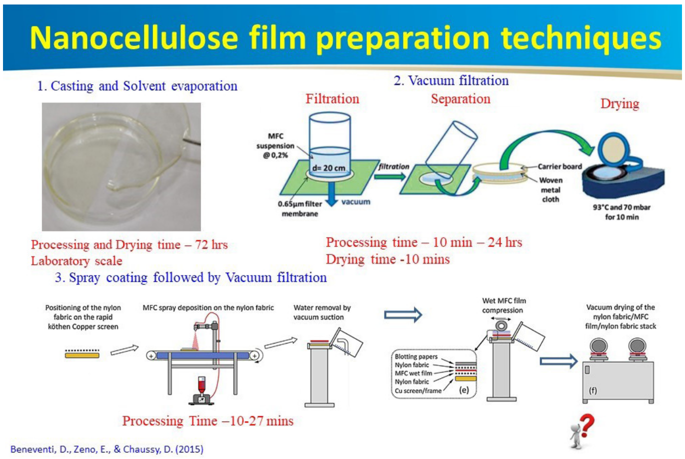


Figure 5: Preparation of free standing nanocellulose films.

Preparation of free-standing nanocellulose films involves several steps. Here is a general procedure that can be followed: Isolation of nanocellulose: Nanocellulose can be obtained from various sources such as wood, bacteria, and plants. The isolation method depends on the source material. Common methods include acid hydrolysis, enzymatic hydrolysis, and mechanical treatment. Acid hydrolysis involves treating the cellulose with acid to break it down into smaller nanofibrils. Dispersion and homogenization: The isolated nanocellulose is then dispersed in a suitable solvent to form a stable suspension. The dispersion can be achieved by mechanical stirring, ultrasonication, or high-pressure homogenization. The purpose is to obtain a uniform suspension of nanocellulose in the solvent. Film casting: The nanocellulose suspension is then cast onto a substrate or a mold to form a thin film. The choice of substrate or mold depends on the desired film properties. Common substrates include glass, silicon wafers, and Teflon-coated surfaces. Alternatively, molds can be used to create structured films with patterned surfaces. Drying: The cast film is dried to remove the solvent and form a solid nanocellulose film. The drying process can be carried out at room temperature, under vacuum, or at an elevated temperature depending on the solvent used. Care should be taken to avoid excessive drying conditions that may cause cracking or shrinkage of the film. Post-treatment: After drying, the nanocellulose film can undergo various post-treatments to enhance its properties. These treatments may include cross-linking, surface modification, or chemical functionalization. Cross-linking agents such as glutaraldehyde or epoxides can be used to improve the mechanical stability and water resistance of the film. Removal from substrate: If a substrate was used during film casting, the nanocellulose film needs to be carefully peeled off from the substrate. This can be done by gently lifting one edge of the film and slowly peeling it off. The resulting nanocellulose film is now free-standing and can be used for various applications. It is important to note that specific details and conditions may vary depending on the specific nanocellulose source, solvents, and desired properties of the film. Therefore, it is recommended to refer to the literature or follow specific protocols established for the particular nanocellulose material and application.

The most common methods for producing free standing nanocellulose films are solvent casting, vacuum filtration and spraying. In casting methods, the nanocellulose suspension was poured into the petridish and allowed to evaporate the water from the cast suspension. The cast film has wrinkles on the surface of the film which affect the uniformity of the film. Vacuum filtration is the conventional method for fabrication of nanocellulose films. In filtration process, the nanocellulose suspension was poured into the column and allowed to drain the water through the filtered mesh under vacuum. The nanofibers were formed as film on the filtered mesh and peeled from the mesh after draining the water and wet film couching with blotting papers. Spraying is an alternative process for developing nanocellulose film via spraying nanocellulose suspension on the fabrics and followed by the water removal from fabric by applying the vacuum.

The spraying process for the fabrication of free-standing nanocellulose films involves the following steps: Preparation of nanocellulose dispersion: Nanocellulose, which is derived from cellulose fibers through mechanical or chemical treatment, is dispersed in a suitable solvent to create a stable nanocellulose dispersion. The solvent used can vary depending on the type of nanocellulose and its compatibility. Spray nozzle selection: A spray nozzle is selected based on the desired droplet size and spraying pattern. The nozzle should be capable of producing a fine mist of nanocellulose dispersion. Substrate preparation: A suitable substrate is prepared for the deposition of the nanocellulose dispersion. The substrate can be a glass slide or any other flat surface that allows easy removal of the deposited film. Spraying process: The nanocellulose dispersion is loaded into a spray gun or airbrush equipped with the selected nozzle. The spray gun is positioned at a specific distance from the substrate surface to ensure uniform and controlled deposition. Adjustment of spraying parameters: The spraying parameters are adjusted according to the desired film thickness and morphology. Parameters such as spray pressure, spray angle, distance from the substrate, and spraying time are optimized to achieve the desired film characteristics. Spraying of nanocellulose dispersion: The nanocellulose dispersion is sprayed onto the substrate surface using the spray gun. The spray gun is moved back and forth, covering the entire substrate area with a thin layer of nanocellulose dispersion. Drying: After the spraying process, the deposited nanocellulose dispersion needs to be dried to remove the solvent. Various drying methods can be employed, including air drying, vacuum drying, or heat drying. The drying conditions should be carefully controlled to prevent cracks or defects in the resulting film. Film detachment: Once the nanocellulose film is dried and firmly attached to the substrate, it can be gently detached from the substrate surface. The free-standing film can then be further processed or used for various applications. By following these spraying steps, free-standing nanocellulose films with controlled thickness and morphology can be fabricated for applications such as flexible electronics, biomedical devices, and packaging materials.

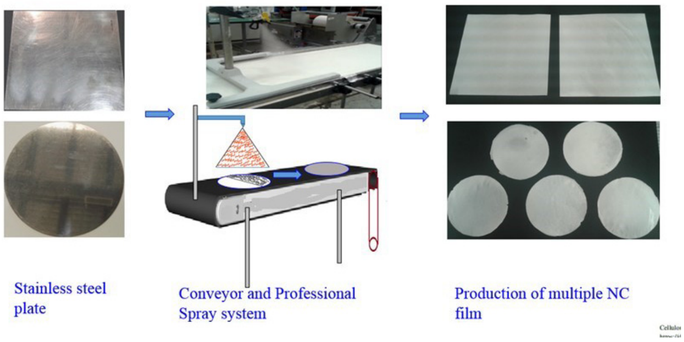


Figure 6: Spraying Process for fabrication of free standing nanocellulose films.

Figure 6 shows the newly developed spraying process for the fabrication of free standing nanocellulose films. In newly spraying process, spraying nanocellulose suspension on the stainless steel plates was attempted. The dried film from the stainless steel plate was peelable easily and the film has two unique surfaces namely rough surface exposed to air and smooth surface exposed to stainless steel plate. The smooth side of the film has roughness of 400nm and used to fabricate various functional materials such as printed electronics and flexible electronics materials.

The operation time for spraying nanocellulose suspension to form 15.9 cm diameter film was less than a minute. It confirms that the spraying process was very rapid in the formation of wet film. The spray coated film was subjected to recyclability studies into a secondary barrier material.

Sustainability in packaging demands using recyclable packaging materials and reducing pollution and carbon emissions. Engineering food packaging materials must include safety and sustainability. On the basis of their very low genotoxicity towards hepatocytes and aquatic organisms, cellulosic fibres have very low toxicity. NC is biodegradable and does not infiltrate or interact with the food chain, like cellulose macrofibers. Nanocellulose may be broken down by microbes in the environment. The tiny size of cellulose nanofibrils exposes it in the environment for microbial activity. Cellulose is a substrate for organisms that express cellulose, such as fungus and bacteria. As a result, nanocellulose has the potential to be a sustainable, biodegradable barrier material. Resources that are abundantly available and renewable can be used to create cellulose nanofibers.

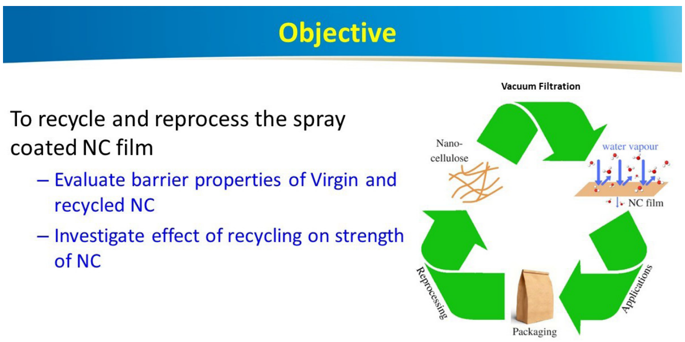


Figure 7: Motivation of the scientific study.

This paper reveals the recyclability of the spray coated nanocellulose films and the barrier performance of the secondary nanocellulose film from recycling of virgin nanocellulose film. In addition to that, the effect of recycling on the mechanical strength of the film was also addressed.

Materials and Methods

The literature has not consistently used the terminology for nanocellulose. In addition to being known as Nanocellulose (NC), it is also known as Nano-Fibrillated Cellulose (NFC), cellulose nano-fibrils, and cellulose micro-fibrils. In this article, we refer to the cellulose nanoparticles utilised generally as NC. NC with a 25% solids concentration was provided by DAICEL Chemical Industries Limited (Celish KY-100S). The cellulose fibrils in DAICEL NC (Celish KY-100S) have an average diameter of 73 nm, a wide range of fibre diameters, a mean length of around 8 m, and an average aspect ratio of 142. By microfibrillating cellulose under high water pressure, DAICEL KY-100S is made. DAICEL nanocellulose's crystallinity index was determined to be 78%. By reducing the initial concentration of 25 wt.% to 1.5 wt.% with deionized water and dissolving for 15,000 revolutions at 3000 revolutions per minute in a disintegrator, NC suspensions were produced.

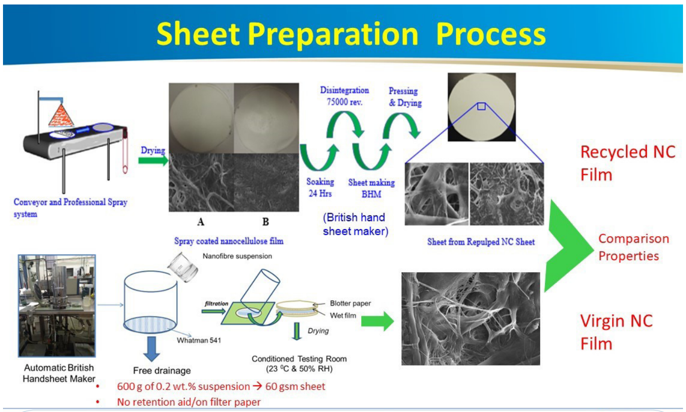


Figure 8: Preparation of recycled nanocellulose sheet from virgin nanocellulose films.

Preparation of Nanocellulose film by Vacuum filtration

According to the scientific literature, the Virgin NC films were produced using the traditional vacuum filtering procedure and a British Handsheet Maker (BHM). A cylindrical container with a 150 mesh filter at the bottom was filled with 600 ml of NC suspension with a 0.2 wt.% concentration, and the solution was filtered to create a wet film on the mesh. Blotting sheets were used to properly separate the wet film, which was then dried at 105°C in a drum dryer for around 10 minutes.

Recycling of Nanocellulose Films

Recently, a spray coating technique for creating NC films on polished, smooth stainless steel surfaces was devised [17]. This technique can spray high-solids NC suspension. We prepared 40 films using this method [17, 18] by spraying onto a smooth stainless steel plate on a moving conveyor at a fixed velocity of 0.32 cm/sec using a Professional Wagner spray system (Model number 117) at a pressure of 200 bar in order to create the large number of films needed for recycling. The elliptical spray jet produced by the type 517 spray tip in a spray system has a beam width of 22.5 cm and an angle of 50°.

The spray distance between the spray nozzle and the round steel plate is 30.01.0 cm. After spraying, the plate's film was allowed to dry for at least 24 hours while being held back at the edges. Following TAPPI standard T205 SP-02, the dried nanocellulose films were divided into 1 cm by 1 cm pieces and immersed in deionized water for 24 hours before being dispersed and suspended in water. The films were dissolved in a disintegrator Model MK III C for 75,000 revolutions at 3000 rpm. The resulting suspension was then transformed into films using the previously described vacuum filtering technique.

Determination of NC fibre diameter

On the silicon chip, a drop of 0.2 weight percent of a recycled or virgin nanocellulose solution was applied, and it was allowed to dry in a sterile laboratory setting. Iridium is layered over the dried suspension on the silicon device. With a voltage of 5 KV, its surface was examined utilising secondary electron mode-II with an FEI Magellan 400 FEGSEM. The images are captured at microns. Image J was used to measure the diameter distribution of the virgin and recycled nanocellulose fibres. The results are given as the average NC diameter within a 95% confidence range. The diameter distribution histograms and micrographs of virgin and recycled nanocellulose fibres are provided in this paper.

Characterization of NC films

Prior to further testing, all prepared virgin and recycled NC films were kept for 24 hours at 23 ^{O}C and 50% RH. Each nanocellulose film's basic weight (g/m2) was determined by dividing its weight by its surface area after it had dried for four hours at 105 °C in the oven. A Thickness Tester Type 21 manufactured by Lorentzen & Wettre AB, Stockholm, Sweden, was used to measure the thickness of the virgin and recycled NC films. Each NC film's thickness was measured fifteen times, with the average value being used. NC film's thickness was determined in accordance with TAPPI T 411, 2015. By dividing basis weight by film thickness, the apparent density of NC films was calculated.

Air permeance of the NC film

An L&W air permeance tester with an operating range of micrns/pa.s. Was used to measure the air permeance of dried NC films. Each NC film's average air permeance rating, calculated from three distinct places, was provided. The air permeance of the films is determined using the TAPPI (Technical Association of the Pulp and Paper Industry) standard T 460.

Evaluation of Water Vapour Permeability of NC films

Using anhydrous Calcium Chloride (CaCl2), Water Vapour Permeability (WVP) was assessed in accordance with the American Society of Testing and Materials (ASTM) standard (E96/E96M-05) technique. In this process, NC film was dried in an air oven for 24 hours at 105 °C. The cups were filled with around 40 g of dry, anhydrous CaCl2, and the NC films were placed on top. $CaCl_2$ within the cups through NC film absorbs water vapour, increasing the weight of the cups. The test sample was weighed at regular intervals. The cups' weight varied with time, and the WVTR was determined using the slope of the relationship between weight and time. At 23°C and 50% Relative Humidity (RH), tests were conducted to measure the water vapour transfer rate. The NC film's Water Vapour Transmission Rate (WVTR) is normalised for thickness and converted to WVP. Three copies of each virgin NC film were tested, and the mean value was recorded. Four distinct recycled NC films were valued and reported.

Evaluation of Uniformity of NC film

The Paper Perfect Formation (PPF) Tester (OpTest Equipment Inc, Canada), which gauges the optical uniformity of light passed through the sample, was used to assess the uniformity of the NC films. In a nutshell, the PPF employs a CCD camera interfaced with 256 grey levels, 65 m/pixel resolution, and a black and white camera based image analyzer. A diffuse quartz halogen light source with IR filters and automated intensity control is used by the analyzer. The PPF divides formation quality into 10 categories using length scales that range from 0.5 mm to 60 mm. The information presented here is the Relative Formation Value (RFV) of each component in relation to the reference film produced by one of the vacuum filtration films. For each condition, three films were examined, and the findings were averaged. If the RFV value is less than 1, it signifies that the NC film tested has poorer optical uniformity than the reference film at that length scale.

Surface Properties of the NC film

By using a Parker surface instrument and optical profiler (Bruker Contour GT-I), the NC films' surface roughness was assessed. To reliably assess RMS of NC film, the raw picture from optical profilometry is processed using Gwyddion 2.49. For both virgin and recycled NC film, the surface topography of the film was examined using the FEI Magellan 400 in secondary electron mode-II.

Mechanical Properties of the film

Using test specimens that were 100 mm long and 15 mm wide and conditioned for 24 hours at 23 °C and 50% RH prior to dry tensile testing in accordance with the Australian/New Zealand Standard AS/NZS 1301.448S-2007, the strength of both virgin and recycled NC films was assessed using an Instron model 5566. At 23°C and 50% RH, all thickness and tensile tests were conducted. The samples underwent testing at a continuous rate of 10mm/min elongation. Tensile strength (expressed in Nm-1) divided by basis weight (expressed in gm-2) was used to compute the tensile index (TI) of the samples. The error bars in the graphs show the standard deviation, and the mean value was derived from six to seven reliable experiments.

Six samples, each 1.5 cm broad and 0.5 cm long, were used for each test to analyse the films' zero span tensile index using a Pulmac Troubleshooter [19]. In a nutshell, each investigation required six samples of NC film. To ensure good jaw alignment under pressure, a sample of NC film was placed in the tester's centre clamping region, and two more samples were stored beneath the clamping jaws' two rear steps. The samples were taken out and replaced after each test. It was decided what the ideal clamping pressures should be before the zero-span testing. For samples of nanocellulose film, a clamping pressure of 70 psi was found to be ideal.

Result and Discussion

The diameter for nanocellulose from virgin and recycled pulp has been evaluated. It has been notified that there are no significant changes in the fibre diameter. Table 1 shows the descriptive statistics analysis of the recycled fibres and virgin fibers. Figure 9 shows the SEM micrograph of the virgin and recycled fibres and the recycling process does not affect the fibre diameter and the fibre morphology and its topography.

Recycling of Nanocellulose

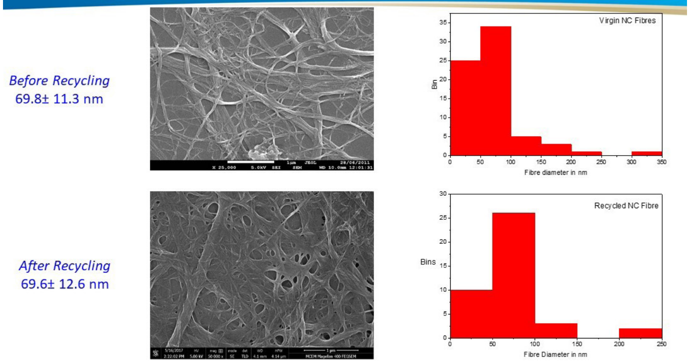


Figure 9: Fibre diameter-Virgin fibres and Recycles fibres.

The diameter of raw nanocellulose was ~70 nm and the effect of recycling on the diameter of nanocellulose fibres is not affected. Table 1 shows that that there is no difference between the virgin fibres and recycled fibres.

Table 1: Results of fibre diameter from statistical analysis.

Before recycling Fibre Diameter	
Mean	0.065321429
Standard Error	0.004326018
Median	0.042
Mode	0.028
Standard Deviation	0.022891133
Sample Variance	0.000524004
Kurtosis	1.655927233
Skewness	1.137471992
Range	0.098
Minimum	0.012
Maximum	0.11
Sum	1.269
Count	28
Confidence Level(95.0%)	0.008876255
After recycling Fibre Diameter	
Mean	0.065550725
Standard Error	0.004735526
Median	0.045
Mode	0.044
Standard Deviation	0.039336234
Sample Variance	0.001547339
Kurtosis	12.79286063
Skewness	2.984723481
Range	0.258
Minimum	0.009
Maximum	0.267
Sum	3.833
Count	69
Confidence Level (95.0%)	0.009449595

Apparent Density

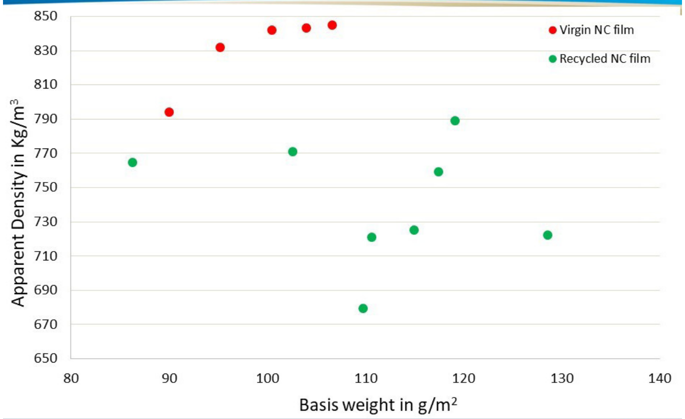


Figure 10: Apparent density of the recycled nanocellulose films and virgin nanocellulose films.

Figure 10 depicts the connection between film density and base weight for all the sheets produced for this investigation. Figure 10 illustrates a shift in apparent density that results from reprocessing. The increased apparent density of NC films made from virgin fibres shows that a less compact structure develops during recycling. It is important to highlight that the recycled film and virgin film were produced using the same vacuum filtering technique, therefore the loss in apparent density during recycling is due to the qualities of the fibre and not the method. Both recycled and virgin NC film have almost the same density, which is independent of basis weight, although recycled NC film has a statistically lower density. The creation of bigger aggregates in the nanocellulose fibrous network, which has prevented the compaction of the fibre network, is likely what has caused the lower density of recycled NC film. Recycled NC films have wider pores as a consequence, which has increased air and water vapour permeability via the pores.

Barrier Properties of Virgin and Recycled NC

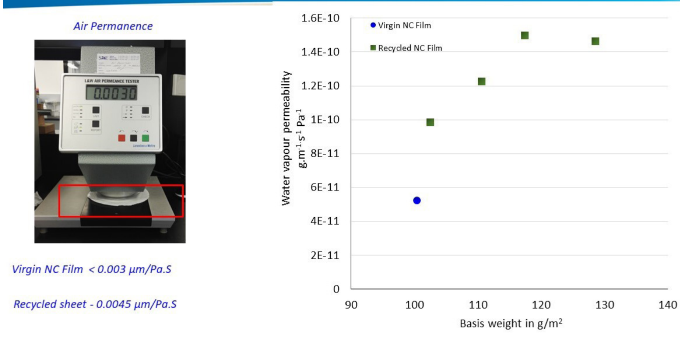


Figure 11: Barrier Potential of Air Permeance and Water vapour permeability of the recycled and virgin nanocellulose films.

Figure 11 reveals the barrier performance of the virgin spray coated NC film and recycled NC film prepared via vacuum filtration. The air permeance of the virgin NC film was reported to be < 0.003µm/Pa.S confirming that the film was impermeable against air and other gaseous substances. It shows that the virgin film was a good packaging material and alternative for synthetic plastics. The air permeance of the recycled NC film was evaluated to be 0.0045 µm/Pa.S nearing the value of 0.003µm/Pa.S. Due to hornification of cellulose nanofibers, the film was bit higher air permeance. For each sample, the Water Vapour Permeability (WVP) was assessed in triplicate. Figure 11 compares the outcomes in relation to film base weight. In comparison to virgin NC films, recycled cellulose films exhibit greater WVP, with an average increase of around 7.5 × 10-11 g.m-1.s-1 Pa-1 (Figure 11).

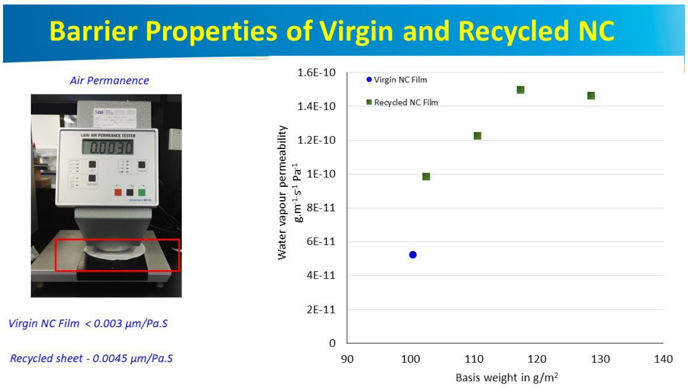


Figure 12

Both the conventional 100 mm test spans, in an Instron, and at zero-span were used to assess the tensile strength of the virgin and recycled NC films. The nanofibre film strength over a very brief span is measured by the zero span tensile index [19]. Figure 12 demonstrated that the recycled film's tensile index was lower than the virgin film for both long and short spans, declining by 30% and 28%, respectively. Due to less effective hydrogen bonding between the strands and their hornification, it is anticipated that recycled conventional cellulose fibres would lose mechanical strength. Weaker inter-fibre connections caused by the cellulose fibres' decreased conformability have an impact on the tensile index of recycled film [20].

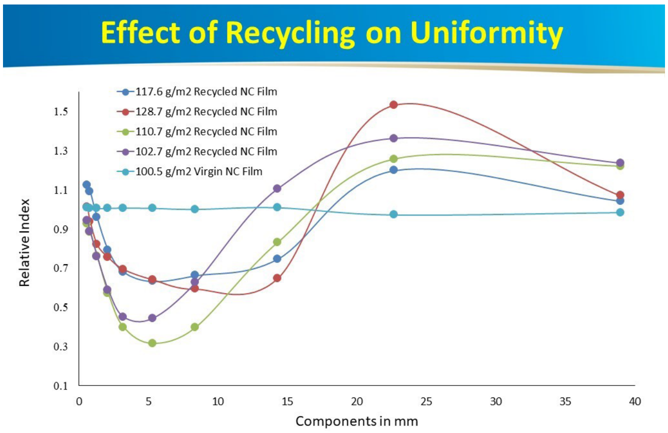
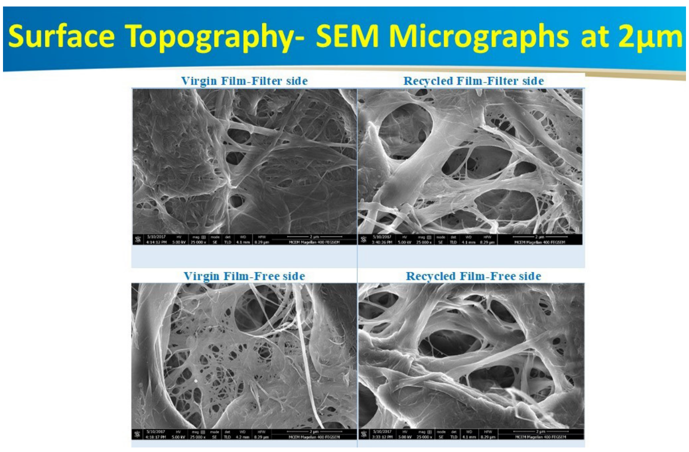
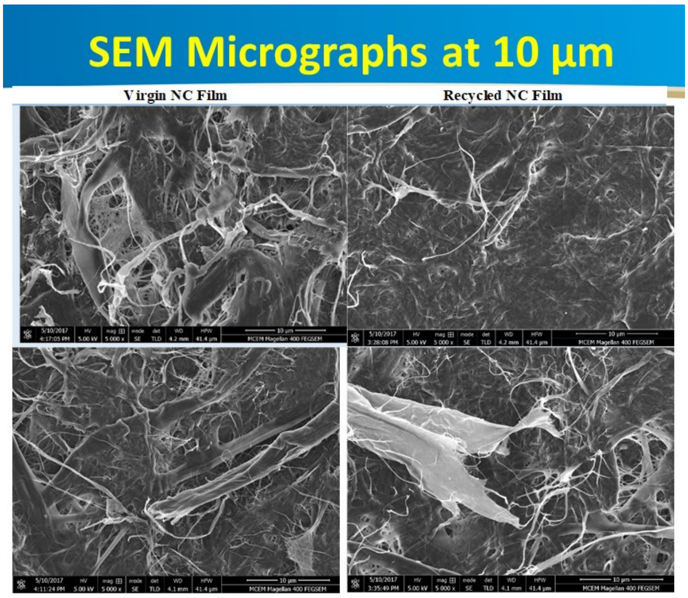


Figure 13

The forming test shows that recycled NC films have better homogeneity than new NC film. Figure 4 further suggests that recycled film is more uniform for areas above 15 mm and less uniform for areas below 15 mm compared to virgin NC film.



With the use of SEM micrographs, the surface morphology, topography, and diameter of the nanocellulose fibres were examined in order to assess the impact of recycling. SEM micrographs of the NC fibres before and after recycling are shown in Figure 6, demonstrating the vast range of fibre sizes in each sample. The nanocellulose had an average diameter of 69.8 11.3 nm prior to recycling and 69.6 12.6 after, with no signs of aggregation in the lower inspection region. Figure 5's SEM micrographs reveal a more compact structure following recycling but no rise in average fibre diameter.



SEM micrographs of the surface topography and morphology of recycled films were analysed and contrasted with those of virgin NC films in Figure 7. The recycled films showed no discernible degradation to the fibre shape or topography. Cellulose fibrils were positioned and dispersed at random. Some bigger holes were seen in the recycled film, which might be the cause of the increased air permeance and water vapour transfer. The supplemental information includes further micrographs of both virgin and recycled NC fibres, as well as NC film made from virgin and recycled NC fibres, ranging in size from 1 to 100 m.

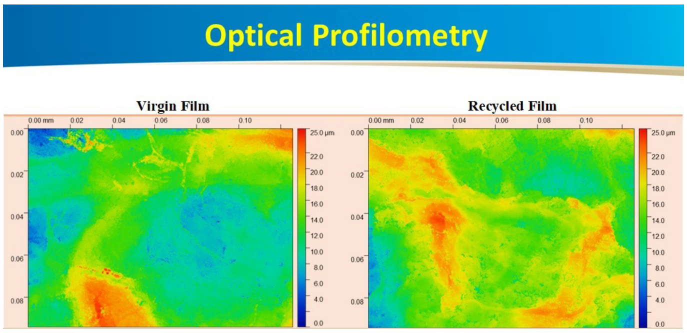


Figure 8 displays the optical profilometry-measured surface roughness of the recycled and virgin NC films. The RMS roughness of the recycled film was determined to be 2624 nm and 3116 nm on the filter side and free side, respectively, exhibiting a similar roughness to the values of 2445 nm on the filter side and 3113 nm on the free side after recycling for an inspection area of 125.7 m x 94.2 m. RMS roughness of 2673 nm and 3751 nm for NC film by vacuum filtering is demonstrated by previously reported data [17]. The supplemental material includes additional photos of optical profilometry taken at a 5X magnification, which show that the surface roughness of virgin and recycled NC film is identical.

Discussion

Recycling has an impact on virgin NC fibres' zero-span strength, strength, WVP, air permeability, and optical homogeneity. This effect is primarily seen at greater inspection areas. The conventional hypothesis of fibre hornification during recycling, which affects the bonding between fibres and primarily lowers the strength of NC film, is unable to explain the change in the characteristics of recycled NC fibres. The best explanation we have is that little flakes remain from some of the agglomeration that was partially broken down during the first film processing. These material flakes are most likely around 100 microns in size. If there are material flakes in the structure that are connected by NC fibres, they won't pack together as well as individual fibres would, which will reduce density, uniformity, and strength at various length scales while simultaneously increasing permeability and only slightly decreasing density. These impacts are limited, though, and recycling has no negative impact on the reprocessed product's applicability-it still has a good barrier performance. It should be mentioned that by maximising the recycling conditions and processing, the recycled film's qualities may be further enhanced. Increasing the disintegration period or utilising well-established chemistry from the paper recycling process to enhance the fibril separation are two potential approaches [21]. Another option is to homogenise the broken fibres; this technique proved successful in dissolving clay agglomerates in a cellulose nanofiber solution [22]. The impacts of the creation of bigger aggregates might also be counteracted by these other mechanisms. Due to a significant increase in the number of small pores in the films, combining virgin and recycled NC fibres may also result in films with improved barrier performance. This study has shown that recycling does not affect fibrils at the microscale, allowing them to maintain their size and structure. There is a little detrimental effect on WVP and air permeability, which is probably caused by the tiny agglomerates. This could have lessened the strength, although hornification and other variables might also be at play.

By comparing recycled NC film with various synthetic and non-recycled cellulose polymers, its potential as a packaging material may be assessed. At around 100 g/m^2, the WVP of virgin NC films is 4.97 × 10-11 g/m.s.Pa, whereas the recycled films' WVP is 9.83 x 10-11 g/m.s.Pa. This is a little higher than WVPs for materials used in plastic packaging. For instance, the following have been reported: 3.41x10-12, 7.54x10-12, 6.78x10-12, 8.75*10-13, and 2.94 *10-13 g/m.s.Pa for Ethylene-vinyl acetate (EVA), Polyamide (PA), Polycarbonate (PC), Low Density Poly-Ethylene (LDPE), and Poly Propylene (PP), respectively [[5], Data taken http://usa.dupontteijinfilms.com]. Higher thickness NC films, however, can compensate for this discrepancy. While ordinary 100 g/m2 films generated in this study have a thickness of roughly 100 microns, the standard thickness of plastics described is between 15 and 25 microns. This results in WVTRs that are lower than those of PA and PC and nearly equivalent to those of EVA. This illustrates the usefulness of recycled goods and the appropriateness of the recycling procedure. The virgin NC films of Cellulose Nanofibrils (CNF) of 8.12 x 10-11 and the acetylated CNF of 6.35*10-11 g/m.s.Pa that were previously described in Rodionova, 2011 [23] likewise had comparable WVP with the recycled NC films.

The impacts of recycling on fibres have an impact on the strength of the recycled films as well; nonetheless, 70% of the film's tensile strength was conserved during the recycling process. This too may be improved even further by the previously mentioned approaches. The existence of undamaged fibres is confirmed by the recycled film micrographs. The recycled film's roughness is comparable to both the rough side of the vacuum-filtered NC film and a film made using vacuum filtration. This is crucial because the network of pores and surface roughness in NC films govern key aspects of barrier performance, and variations in roughness might result in different behaviours related to the wettability of NC surfaces [24].

Furthermore, the recycling method employed here is chemical-free, environmentally benign, and straightforward. The ease of upscaling for recycling operations, as well as the effectiveness and simplicity of reprocessing—that is, soaking and disintegration-also highlight the special benefits gained from this method. In situations where recycling and reprocessing are either impossible or extremely complex, recycled NC film can be used as a substitute to synthetic polymers.

Plastics made from fossil fuel byproducts, like LDPE, are inexpensive and simple to shape into the proper shape. These plastics pose a major risk to the ecology since they are neither renewable nor biodegradable. 322 million tonnes of plastic are manufactured year, and because they do not biodegrade, a significant portion of them end up as garbage in the environment [25]. The economic benefits of plastic packaging will be diminished as a result of recent regulations the European Union implemented to limit the use of single-use plastics in applications like packaging [26,27]. Plastics made from fossil fuels might be replaced by biopolymers. Biopolymers like chitosan and starch, however, lack sufficient strength and noticeable barrier efficacy. Consequently, NC fibres are being actively researched as barrier layer substitutes for traditional polymers due to their superior strength and barrier performance [28].

According to a preliminary examination of energy consumption comparing vacuum filtering and spray coating, vacuum filtration needs 0.56 MJ/kilogramme, whereas spray coating uses 0.15 MJ to spray 1 kilogramme of dry NC film. The energy needed to spray the film is comparable to what's needed to make a typical Low-Density Polyethylene (LDPE) film.

Ten times as much energy is needed to produce nanocellulose as it is for conventional packaging films [29,30], though extensive research is being done to reduce this energy usage. Furthermore, it should be mentioned that estimates of energy consumption have frequently been derived using laboratory scale production, and that full scale NC production is still in its early stages of development. As production facilities are scaled up, significant energy savings may be anticipated, according to recent analysis [31].

It will also require a lot more water and land to grow biomass for nanocellulose than it does to produce polyolefin products. This specific comparison makes it easy to conclude that conventional packaging films are more sustainable and that the trade-off between packaging made from petroleum and packaging made from biofuels may only result in the conservation of non-renewable resources. Only two thirds of packaging films, however, can be recycled; the remaining portion is either disposed of in a landfill or released into the environment [32].

In the near future, packaging waste will overflow landfills and cause serious environmental problems. The development of work to incorporate the effects of plastic waste into life-cycle analyses is still in its very early phases [32]. It's also interesting to note that compared to the energy needed to produce these virgin films, recycling polyolefin films like LDPE uses more than half as much energy [33]. However, compared to the energy emissions from its production, recycling nanocellulose requires very little energy [29]. When compared to traditional packaging films, nanocellulose is easier to recycle, making it potentially more environmentally friendly and sustainable when looking at end of life analyses. For this reason, it deserves active development.

Conclusion

By using a standard approach, virgin NC films with great uniformity and low permeability were produced. Virgin NC films were recycled and reformed into films utilising conventional paper testing procedures. The findings demonstrate that tiny flakes remain after part of the agglomeration from the original film production has not completely broken down. Flakes that were partially made of did not pack together as well as individual fibres, which decreased strength, homogeneity, and density. Nonetheless, the recycled NC barrier materials showed very minor decreases in barrier effectiveness and kept around 70% of their tensile strength, making them suitable for the majority of synthetic packaging applications. Nowadays, nanocellulose sheets offer a viable substitute for traditional packaging and may be easily recycled and repurposed as a possible barrier material. By substituting non-renewable, environmentally harmful synthetic plastic packaging and laminates with a substance that may be recycled straight into a barrier material or utilised in the traditional paper recycling process, this technique promotes global sustainability.

References

1. Smithers Pira. The Future of Global Packaging to 2020. 2015 14 December. 2015. Available from: http://www.smitherspira.com/industry-market-reports/packaging/the-future-of-global-packaging-markets-to-2020.
2. Davis G, JH Song. Biodegradable packaging based on raw materials from crops and their impact on waste management. Industrial Crops and Products. 2006; 23(2): 147-161.
3. Tukker A. Plastic waste-feedstock recycling, chemical recycling and incineration. Rapra Rev Rep. 2002; 13(4): 1-10.
4. Bugnicourt E, et al. Processing and Validation of Whey-Protein-Coated Films and Laminates at Semi-Industrial Scale as Novel Recyclable Food Packaging Materials with Excellent Barrier Properties. Advances in Materials Science and Engineering. 2013; 2013(11): 10.
5. Bhunia K, et al. Migration of Chemical Compounds from Packaging Polymers during Microwave, Conventional Heat Treatment, and Storage. Comprehensive Reviews in Food Science and Food Safety. 2013; 12(5): 523-545.
6. Lau OW, SK Wong. Contamination in food from packaging material. Journal of Chromatography A. 2000; 882(1-2): 255-270.
7. Lickly TD, KM Lehr, GC Welsh. Migration of styrene from polystyrene foam food-contact articles. Food and Chemical Toxicology. 1995; 33(6): 475-481.
8. Hubbe MA, et al. Nanocellulose in Thin Films, Coatings, and Plies for Packaging Applications: A Review. BioResources. 2017; 12(1): 2143-2233.
9. Tharanathan RN. Biodegradable films and composite coatings: past, present and future. Trends in Food Science & Technology. 2003; 14(3): 71-78.
10. Azeredo HMC, MF Rosa, LHC Mattoso. Nanocellulose in bio-based food packaging applications. Industrial Crops and Products. 2017; 97: 664-671.
11. Aulin C, M Gällstedt, T Lindström. Oxygen and oil barrier properties of microfibrillated cellulose films and coatings. Cellulose. 2010; 17(3): 559-574.
12. Bras J, S Saini. 8-Nanocellulose in functional packaging A2 - Jawaid, Mohammad, in Cellulose-Reinforced Nanofibre Composites, S. Boufi and A.K. H.P.S, Editors. 2017, Woodhead Publishing. 2017; 175-213.
13. Abdul Khalil, HPS, et al. A review on nanocellulosic fibres as new material for sustainable packaging: Process and applications. Renewable and Sustainable Energy Reviews. 2016; 64: 823-836.
14. Varanasi S, R He, WJ Batchelor. Estimation of cellulose nanofibre aspect ratio from measurements of fibre suspension gel point. Cellulose. 2013; 20(4): 1885-1896.
15. Raj P, et al. Gel point as a measure of cellulose nanofibre quality and feedstock development with mechanical energy. Cellulose. 2016; 23(5): 3051-3064.
16. Varanasi S, WJ Batchelor. Rapid preparation of cellulose nanofibre sheet. Cellulose. 2013; 20(1): 211-215.
17. Shanmugam K, et al. Rapid preparation of smooth nanocellulose films using spray coating. Cellulose. 2017; 1-8.
18. Shanmugam K, et al. Flexible spray coating process for smooth nanocellulose film production. Cellulose. 2018.
19. Varanasi S, HH Chiam, W Batchelor. Application and interpretation of zero and short-span testing on nanofibre sheet materials. Nordic Pulp and Paper Research Journal. 2012; 27(2): 343.
20. Delgado-Aguilar M, et al. Are Cellulose Nanofibers a Solution for a More Circular Economy of Paper Products? Environmental Science & Technology. 2015; 49(20): 12206-12213.
21. M Hietala K, Varrio L, Berglund J, Soini, K Oksman. Potential of municipal solid waste paper as raw material for production of cellulose nanofibers. Waste Management. 2018; 80: 319-326.
22. U Garusinghe S, Varanasi VS, Raghuwanshi G Garnier, WJ Batchelor. Nanocellulose-Montmorillonite Composites of Low Water Vapour Permeability. Colloids & Surfaces A-Physicochemical & Engineering Aspects. 2018; 540: 233-241.
23. Rodionova G, Lenes M, Eriksen Ø, Gregersen Ø. Surface chemical modification of microfibrillated cellulose: improvement of barrier properties for packaging applications. Cellulose. 2011; 18(1): 127-134.
24. Samyn, Pieter. Wetting and hydrophobic modification of cellulose surfaces for paper applications. Journal of Materials Science. 2013; 48(19): 6455-6498.
25. Milios, Leonidas, Aida Esmailzadeh Davani, Yi Yu. Sustainability Impact Assessment of Increased Plastic Recycling and Future Pathways of Plastic Waste Management in Sweden. Recycling. 2018; 3(3): 33.
26. European Commission, Single use of Plastics. 2018. Available from: https://ec.europa.eu/commission/news/single-use-plastics-2018-may-28
27. European Commision, Technical report on. A european strategy for plastics in a circular economy. 2018. Available from: http://ec.europa.eu/environment/circular-economy/pdf/plastics-strategy-brochure.pdf
28. Kim, Joo-Hyung, et al. Review of nanocellulose for sustainable future materials. International Journal of Precision Engineering

and Manufacturing-Green Technology. 2015; 2: 197-213.

29. Li, Qingqing, Sean McGinnis, Cutter Sydnor, Anthony Wong, and Scott Renneckar. “Nanocellulose life cycle assessment.” ACS Sustainable Chemistry & Engineering. 2013; 1,8: 919-928.

30. Jackson, Sue, T Bertényi. “Recycling of plastics.” ImpEE Project, Department of Engineering, University of Cambridge. 2006. Available from; http://wwwg.eng.cam.ac.uk/impee/topics/RecyclePlastics/files/Recycling%20Plastic%20v3%20PDF.pdf.

31. Piccinno F, Hischier R, Seeger S, Som C (2018) Predicting the environmental impact of a future nanocellulose production at industrial scale: Application of the life cycle assessment scale-up framework Journal of Cleaner Production. 2018; 174: 283-295. doi:https://doi.org/10.1016/j.jclepro.2017.10.226

32. Rydz J, Musioł M, Zawidlak-Węgrzyńska B, Sikorska W. Present and Future of Biodegradable Polymers for Food Packaging Applications. In Biopolymers for Food Design. 2018; 431-467.

33. Choi, Bulim, Seungwoo Yoo, Su-il Park. Carbon Footprint of Packaging Films Made from LDPE, PLA, and PLA/PBAT Blends in South Korea. Sustainability.2018; 10.7: 2369.