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Fabrication of Free Standing Nanocellulose Film via Spray Coating and its Biomedical Application: A Review

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Abstract: Spraying nanocellulose onto films provides a quick and scalable way to create free-standing films with exceptional consistency and customizable thickness. This method increases the application of nanocellulose films in various industries and satisfies the requirements of large-scale production. In the field of biomedicine, spray-coated free-standing nanocellulose films hold great promise for applications such as drug delivery, tissue engineering, wound healing, device coatings, and biosensing. They are excellent nanomaterials for a variety of biomedical applications due to their special qualities, including biocompatibility, high mechanical strength, porous structure, large surface area, and adaptability. This paper reviewed the detailed exposure of the spray coating process of nanocellulose suspension onto free-standing films and its biomedical applications.

Keywords: Spraying; Nanocellulose Film; Free Standing Film; Thickness; Basis weight

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1. Introduction

Spraying nanomaterials is a versatile technique with a wide range of applications across various fields. It involves applying nanoparticles ranging from 1–100 nm onto a surface using a spray gun. Nanoparticles exhibit unique properties due to their small size, making them valuable for various purposes. There are 4 common methods for spraying nanomaterials. **Spray coating** involves suspending nanoparticles in liquid and then spraying the suspension onto a target surface. The liquid evaporates, leaving behind a thin film of nanoparticles. This method is suitable for coating various materials, including metals, polymers, and textiles. **Aerosol spraying** uses compressed gas to propel dry nanoparticles out of a nozzle, creating a fine mist that can be deposited onto surfaces. This method is often used for applying nanoparticles to delicate surfaces or those with complex geometries. **Electrospraying** uses an electric field to atomize a liquid suspension of nanoparticles, creating charged droplets that are deposited onto a grounded surface. This method offers precise control over the size and distribution of deposited nanoparticles. **Flame spraying uses** a high-temperature flame to melt and atomize a feedstock of nanoparticles or their precursors. The molten droplets are then propelled onto a substrate, where they solidify to form a coating. This method is suitable for applying high-melting-point materials.

Spraying nanomaterials offers several advantages over other deposition methods. Spraying can produce thin, uniform films of nanoparticles over large areas. The thickness and composition of the deposited film can also be controlled by adjusting the spraying parameters. Furthermore, spraying is relatively inexpensive. However, it is important to consider the potential risks associated with spraying nanomaterials. Airborne nanoparticles can be easily inhaled, potentially causing respiratory problems. Nanoparticles can also contaminate soil and water, and some are flammable or explosive. Hence, it is important to handle nanomaterials with caution and follow appropriate safety protocols. Overall, spraying of nanomaterials is a promising technique with the potential to revolutionize various industries. Nonetheless, it is crucial to carefully consider the safety risks and environmental implications before implementing this technology.

1.2. Spraying nanofibers

Spraying nanofibers, while related to spraying nanomaterials in general, presents a whole other level of complexity and nuance and is presented with 4 techniques.

(1) Electrospinning: This is the most frequently used technique for fabricating nanofibers, which entails utilizing an electric field to pull a charged polymer solution through a small needle, elongating it into extremely thin fibers that harden upon reaching a collection surface. Electrospinning offers precise control over fiber diameter, orientation, and morphology.

(2) Direct spray nebulization: This technique uses compressed air to atomize a polymer solution into a fine mist, with subsequent solvent evaporation leading to nanofiber formation. It is faster than electrospinning but offers less control over fiber properties.

(3) Spray-drying of nanofiber suspensions: This method involves spray-drying a suspension of pre-produced nanofibers in a liquid carrier. The rapid drying process creates a dry powder of entangled nanofibers, which is suitable for further processing or direct application.

(4) Airbrush spraying of nanofiber slurries: This technique uses an airbrush to spray a concentrated slurry of nanofibers onto a surface. It is suitable for creating thicker mats or coatings and offers flexibility in design but requires careful optimization of the slurry viscosity and spraying parameters.

1.3. Applications of sprayed nanofibers

The spray-coated nanofiber films have many applications.

(1) Filtration membranes: Nanofiber mats formed by spraying offer excellent air and liquid filtration with enhanced efficiency and selectivity due to their high surface area and adjustable pore size.

(2) Drug delivery and tissue engineering: Nanofibers can be loaded with bioactive agents and used as scaffolds for tissue regeneration or controlled drug release applications.

(3) Sensors and electronics: Sprayed nanofibers can be functionalized with conductive or catalytic materials, finding applications in flexible electronics, energy harvesting, and biosensing.

(4) Protective coatings: Spraying nanofibers can create hydrophobic, superhydrophobic, or self-cleaning surfaces with applications in textiles, marine coatings, and anti-corrosion protection.

2. Challenges and considerations

There are many challenges in the spraying process and should be considered in the fabrication of free-standing nanomaterial films. It can be challenging to maintain the fiber diameter, distribution, and orientation, especially for large-scale applications. It is crucial to choose environmentally friendly solvents and ensuring proper ventilation during spraying are crucial concerns. Lastly, it is crucial to consider the long-term stability and maintain the desired functionalities of the nanofibers based on the specific application.

3. Spraying cellulose nanofibres

Spraying cellulose nanofibers (CNFs) is a versatile technique used to deposit thin films or coatings onto various substrates. CNFs, with their unique properties like high strength, flexibility, and biodegradability, find applications in diverse fields like filtration, electronics, energy storage, and biomedicine. CNFs can be derived from various sources like wood pulp, bacterial cellulose, or tunicate cellulose. CNFs are dispersed in a liquid, typically water, at a desired concentration (usually 1%–5%). Additives like polymers or surfactants might be used to stabilize the suspension and prevent CNF aggregation. CNFs can be sprayed via airbrush techniques. A handheld airbrush provides precise control over the spraying pattern and film thickness. CNFs can also be sprayed via electrostatic spraying where charged CNF particles are attracted to the grounded substrate to ensure uniform deposition. Pressure spraying allows for a pressurized stream of CNF suspension for larger areas or faster coating.

3.1. Factors affecting the spraying process

Higher concentrations and smaller CNFs generally lead to denser films. Spraying parameters such as nozzle size, pressure, and distance to the substrate also influence film thickness and uniformity. Surface roughness and wettability of the substrate can affect the adhesion and morphology of the CNF film.

Sprayed CNF films are often used for barrier coatings as they offer excellent barrier properties against gases, liquids, and even flames. Adding CNFs to composites can enhance their strength, stiffness, and thermal stability. Besides that, CNFs are used in electrodes for batteries and super-capacitors due to their high surface area and conductivity. They are also used in biomedical applications for wound healing, drug delivery, and tissue engineering due to their biocompatibility and biodegradability.

4. Common methods for the fabrication of free-standing nanocellulose films

There are several common methods used for the fabrication of free-standing CNFs.

(1) Solvent casting: In this process, NC is mixed in a solvent, usually water or a combination of water and organic solvents, to create a liquid mixture. This mixture is then poured onto a surface, like a glass plate or petri dish, and the solvent is left to gradually evaporate, resulting in a solid nanocellulose film.

(2) Spin coating: Spin coating involves depositing a liquid solution or suspension of nanocellulose onto a rotating substrate, commonly a silicon wafer. The rotation spreads the solution uniformly across the substrate, and the solvent is evaporated through spinning, resulting in a thin, homogeneous CNF.

(3) Filtration: In this method, an NC suspension is poured onto a porous filter membrane, such as a cellulose acetate or polycarbonate filter. The liquid component of the suspension passes through the filter, leaving an NC film on the surface of the membrane. The film can then be carefully peeled off as a free-standing membrane.

(4) Layer-by-layer assembly: Layer-by-layer assembly involves the sequential deposition of NC layers onto a substrate using electrostatic interactions or other binding forces. Each layer is formed by immersing the substrate alternately into solutions of oppositely charged NC or other polyelectrolytes. The process is repeated until the desired thickness of the free-standing nanocellulose film is built up.

These methods offer different advantages and can be adapted based on specific requirements such as desired film thickness, porosity, and surface morphology. They enable the fabrication of free-standing CNFs with various applications in areas such as packaging, biomedical engineering, and energy storage. However, the above methods are slow in the formation of the film and time-consuming process and laborious procedure. Hence, a rapid process for the fabrication of free-standing CNF is required.

5. Criteria for fabrication of free-standing NC films

Current techniques for creating standalone NC films are inefficient and cannot be easily expanded to meet the needs of large-scale production. In contrast to the rapid production rates of plastic films and synthetic coatings on paper using extrusion methods, the process of making CNFs is significantly slower. To address the growing demand for these films and enhance their applications across different industries, it is essential to develop faster and more adaptable methods for producing barrier coatings on paper and NC films.

Development should focus on boosting film qualities with a quick preparation technique to ensure consistent uniformity similar to traditional methods like vacuum filtration. The thickness and weight of the NC film can be modified by altering the processing method and concentration of suspension without extending the operation duration. A free-standing film can be prepared in one step to eliminate the need for additional processing like removing water, vacuum drying, and couching the wet film.

The fabrication of free-standing nanocellulose films involves several critical criteria to ensure the desired properties and performance of the films. These criteria can be categorized into the following aspects:

(1) Material selection: (a) Type of Nanocellulose: Choosing the appropriate type of nanocellulose (e.g., cellulose nanocrystals (CNCs), cellulose nanofibrils (CNFs), or bacterial nanocellulose) based on the desired properties of the final film; (b) Purity and quality: Ensuring high purity and quality of the nanocellulose to avoid impurities that could affect film properties.

(2) Preparation methods: (a) Homogeneous dispersion: Achieving a uniform and stable dispersion of nanocellulose in the solvent to avoid aggregation and ensure consistent film properties; (b) Sonication and stirring: Using techniques like sonication and mechanical stirring to aid in the dispersion of nanocellulose.

(3) Casting and drying techniques: (a) Casting methods: Selecting an appropriate casting method such as vacuum filtration, solvent casting, or doctor blade coating to form the film; (b) Drying conditions: Controlling drying conditions (temperature, humidity, drying rate) to avoid defects like cracking or warping in the films.

(4) Structural and mechanical properties: (a) Film thickness: Controlling the thickness of the film, which can affect mechanical properties and transparency; (b) Mechanical strength: Ensuring the film has sufficient tensile strength and flexibility for the intended application; (c) Surface smoothness: Achieving a smooth surface finish to enhance optical clarity and reduce surface roughness.

(5) Chemical and physical modifications: (a) Surface modification: Applying chemical treatments to modify the surface properties of nanocellulose, such as hydrophobicity or functionalization with specific groups; (b) Composite formation: Incorporating other materials (polymers, nanoparticles) to enhance specific properties like barrier performance or mechanical strength.

(6) Performance characteristics: (a) Barrier properties: Evaluating the film's ability to act as a barrier against gases, moisture, and other substances; (b) Optical properties: Ensuring the film has the desired transparency or opacity based on its application; (c) Thermal stability: Assessing the thermal stability of the film to ensure it can withstand the required processing and operating conditions.

(7) Environmental and safety considerations: (a) Biocompatibility: Ensuring the film is biocompatible if intended for biomedical or food packaging applications; (b) Environmental impact: Considering the environmental impact of the fabrication process and the biodegradability of the final product.

(8) Characterization and testing: (a) Microscopy: Using techniques like scanning electron microscopy (SEM) or atomic force microscopy (AFM) to analyze the film's morphology; (b) Mechanical testing: Conducting tensile tests to measure mechanical properties; (c) Spectroscopy: Employing techniques like Fourier transform infrared spectroscopy (FTIR) or X-ray diffraction (XRD) to analyze chemical composition and crystallinity.

By carefully considering these criteria, researchers and manufacturers can optimize the fabrication process to produce high-quality, free-standing nanocellulose films with tailored properties for specific applications.

6. Proof of concept of spray coating

To create a wet NC film, researchers proposed the concept of applying nanocellulose suspension onto a smooth metal surface through spray deposition. Research has shown that when nanocellulose is sprayed onto the surface, the resulting films mimic the roughness of the base surface [1,2]. Consequently, the film's surface roughness can be adjusted to create a smooth surface. The method's need for speed poses the query: "Is it possible to scale up the process for continuous production of NC films like synthetic plastic films?" The process should be able to adjust the bulk and mechanical characteristics of the film during the spraying process. One method that can meet these needs and produce NC films is spraying [3].

Spraying cellulose nanofibers (CNFs) onto stainless steel plates involves a combination of the right techniques and conditions to ensure a uniform and effective coating. Atomization in the spraying process is a critical phenomenon that involves breaking up a liquid into fine droplets. This process is essential in various applications, including coating, painting, fuel injection, and agricultural spraying. **Figure 1** reveals the atomization process during the spray coating process.

Here's a detailed look at the criteria and mechanisms involved in atomization in the spraying process:

(1) Mechanisms of atomization: (a) Pressure atomization: High-pressure liquid is forced through a nozzle, causing it to break into droplets due to shear forces; (b) Air-assisted or airless atomization: A stream of liquid is introduced into a high-velocity air stream, breaking the liquid into droplets; (c) Ultrasonic atomization: High-frequency vibrations create capillary waves on the liquid surface, leading to droplet formation; (d) Rotary atomization: A spinning disk or cup throws the liquid outward by centrifugal force, forming droplets.

(2) Key parameters influencing atomization: (a) Liquid properties: (i) Viscosity: Higher viscosity liquids require more energy to atomize and generally produce larger droplets; (ii) Surface tension: Lower surface tension facilitates easier breakup of the liquid into finer droplets; (iii) Density: The density of the liquid can affect the droplet size and distribution; (b) Nozzle design: (i) Orifice size: Smaller orifices generally produce finer droplets, but may require higher pressures; (ii) Spray angle: The angle of the spray affects the coverage area and droplet distribution; (iii) Material and geometry: The design and material of the nozzle affect its durability and performance; (c) Operational conditions: (i) Pressure: Higher pressures typically result in finer atomization but may require more robust equipment; (ii) Flow rate: The rate at which the liquid is fed into the nozzle affects the spray characteristic; (iii) Air assistance: In air-assisted systems, the velocity and pressure of the air stream are crucial for droplet formation.

(3) Droplet characteristics: (a) Size distribution: Atomization should achieve a uniform droplet size distribution for consistent application; (b) Droplet velocity: The velocity at which droplets are ejected can impact their ability to adhere to surfaces and their overall spread.

(4) Applications and requirements: (a) Coating: (i) Uniform coverage with fine droplets to avoid runs and ensure smooth finishes; (ii) Controlled droplet size for different coating thicknesses and textures; (b) Fuel injection: (i) Fine atomization for efficient combustion and reduced emissions; (ii) Consistent droplet size for optimal air-fuel mixture; (c) Agricultural spraying: (i) Appropriate droplet size to ensure coverage without drift; (ii) Minimized wastage and environmental impact.

(5) Characterization and measurement: (a) Spray pattern analysis: Using high-speed cameras and laser diffraction to analyze droplet size distribution and spray pattern; (b) Viscosity and surface tension measurements: Ensuring the liquid properties are within the optimal range for the chosen atomization method; (c) Flow rate and pressure monitoring: Ensuring consistent operational conditions for repeatable atomization results.

In conclusion, effective atomization in the spraying process requires careful consideration of the liquid properties, nozzle design, and operational conditions. By optimizing these factors, it is possible to achieve the desired droplet characteristics for various applications, ensuring efficient and uniform application of the sprayed material.

A smooth, polished stainless-steel plate was utilized as the surface for spraying a nanocellulose suspension. The plate's bottom surface was exceptionally sleek with minimal scratches. The experimental arrangement for testing the nanocellulose spraying on the stainless-steel plate is illustrated in **Figure 2**. To store the plate post-spraying, the setup includes a professional spray system, a conveyor, and seating arrangements. Nanocellulose (NC) from DAICEL Chemical Industries Limited (Celish KY-100S) was used to create free-standing films. The NC sample was prepared by diluting the original 25% concentration with distilled water and mixing it for 15,000 revolutions in a disintegrator. The NC suspension's viscosity was determined using the flow cup method, which measures the fluid flow through an orifice to provide a relative measurement of kinematic viscosity, with results expressed in DIN-Seconds [2].

In this experiment, a circular stainless-steel plate placed on a moving conveyor was coated with nanocellulose. The conveyor speed and the concentration of the NC suspension were adjusted, as these factors could influence the properties of the resulting nanocellulose films. The process involved spraying a nanocellulose suspension onto the plate at a consistent speed, with the suspension concentration ranging from 0.25–2.00 wt.%. By varying the conveyor speed, more NC could be applied at slower speeds, resulting in thicker and heavier films. This method allows for tailoring the characteristics of the NC films to meet specific requirements [2].

There are many hidden factors in the process of spray coating and the experimental setup that need to be considered to create consistent and high-quality NC films. Atomization is a crucial step in spraying fibers onto the surface. The spray nozzle orifice and the viscosity of the liquid to be sprayed determine the atomization process. The nozzle type and orifice diameter both affect how a spray jet forms. There are three types of spray patterns: elliptical, rectangular, and circular. The spray system's fan types, orifice positions, and spray gun positions can all be used to create these spray patterns [4]. The NC suspension's viscosity should fall within the sprayable range to develop films, excluding equipment issues. For the best spraying performance in this instance, the NC suspension's fiber content was adjusted from 0.25%–2.00%. The solid content of the nanocellulose suspension determines how sprayable the suspension is [5].

A stainless-steel plate placed on a conveyor belt with adjustable speed was coated with an NC suspension by using a Wagner Professional spray system (Model No. 117) at a pressure of 200 bar. The suspension was sprayed onto the plate using a type 517 spray tip, creating an elliptical spray jet with a width of 22.5 cm and an angle of 50°. The spray distance, which is the vertical distance between the spray nozzle and the plate, was maintained at 30.0 ± 1.0 cm. The conveyor's speed during spraying was set at 0.32 cm/sec, and the Wagner spray system was operated for 30 seconds to ensure a uniform coating of NC on the plate. This process was allowed to stabilize throughout the spraying operation [2].

After applying an NC spray coating onto a base material like stainless steel, the wet film on the surface was dried in a typical laboratory environment. It is recommended to use a fume hood during the drying process to help facilitate the film's quick drying. Typically, it takes over 24 hours for the wet NC films to dry at a temperature of 25 ℃. The dried film was then stored at 23°C and 50% relative humidity to evaluate its mechanical and barrier properties. To compare the effectiveness of the novel spraying technique, the spray-coated film was contrasted with the current technique. Vacuum filtration is commonly used for creating NC films, and the film produced through this method was found to be a suitable match for the spray-coated NC film [2].

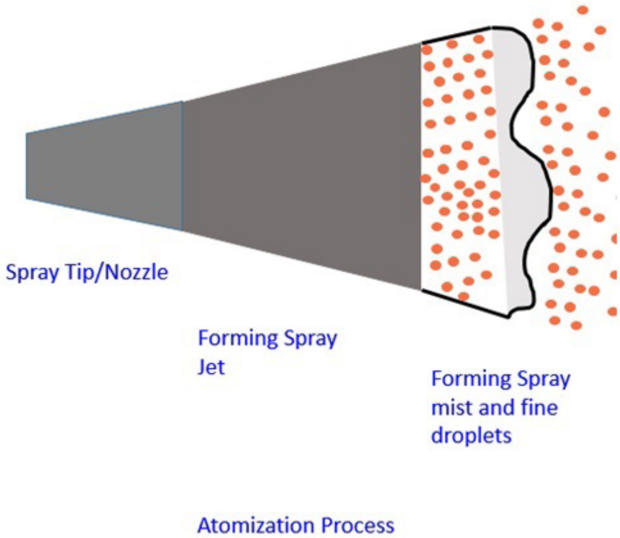

Figure 1. The function of a spray gun for spraying NC fibers of droplets [6]

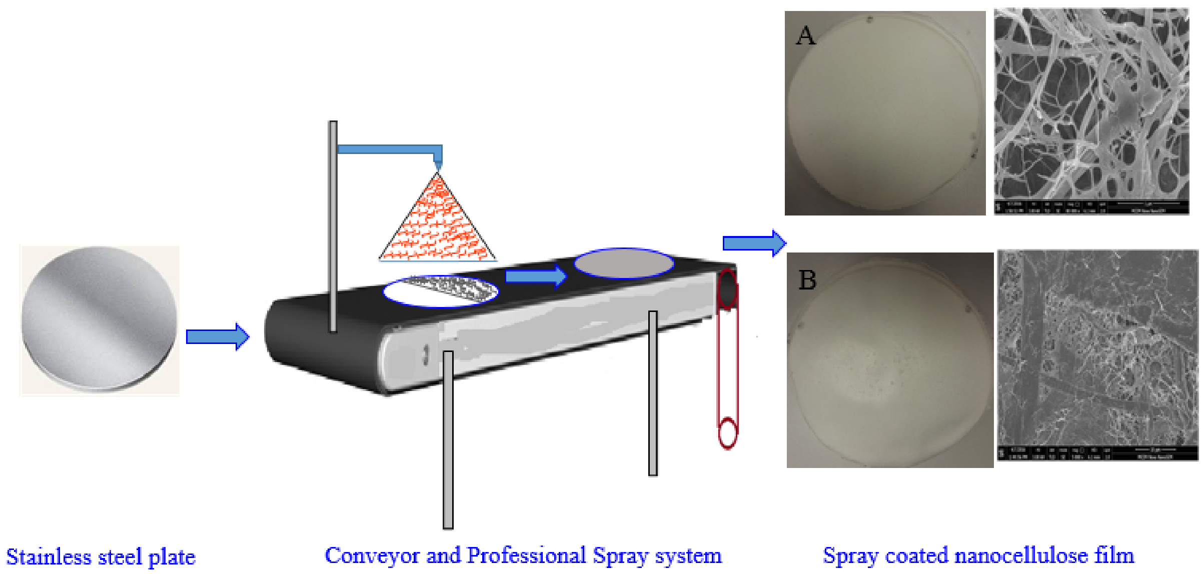

Figure 2. Proof of concept for spray coating for the production of nanocellulose films [2]

NC films are typically made using vacuum filtration as a standard method, which is used as a comparison to spray-coated NC films. The vacuum filtration process involves filling a cylindrical container with a 125-mesh filter at the bottom with an NC suspension (0.2%). The mixture is then filtered until a wet film forms on the mesh, which is carefully separated and dried in a drum dryer at 105°C for 10 minutes. This film serves as a reference for comparing the thickness and uniformity of spray-coated films. The operation time for forming the wet film in the vacuum filtration method is less than a minute on a laboratory scale. In contrast, spray-coating CNFs require a very short operating time and minimal water. The spray-coating process allows for customization of the basis weight and thickness of the films by adjusting the concentration of the NC suspension. However, one limitation of the process is the longer drying time (seconds), which can be addressed by utilizing waste heat or infrared radiation drying methods [7].

7. Spray-coated CNFs

One possible nanomaterial that could be used in place of synthetic plastics is CNFs [8]. Owing to the enormous demand for NC films as a synthetic plastic substitute, a quick and adaptable process was needed to support large-scale production. Spraying is a quick and adaptable method for making free-standing NC films [7]. This technique involves producing a unique nanocellulose film with two separate surfaces by applying an NC suspension onto a smooth metal surface, such as stainless steel [3]. A free surface, also known as a rough surface, is a surface exposed to air. Spraying the film onto the metal surface allows it to stick there, peels off with ease, and mimics the smoothness of the metal side's surface. Consequently, the metal side of the NC film was extremely smooth [2,9].

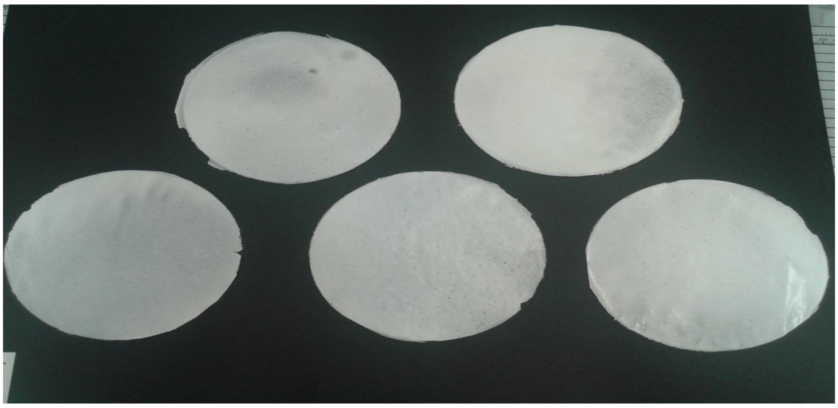

Figure 3. Spray-coated CNFs that are circular

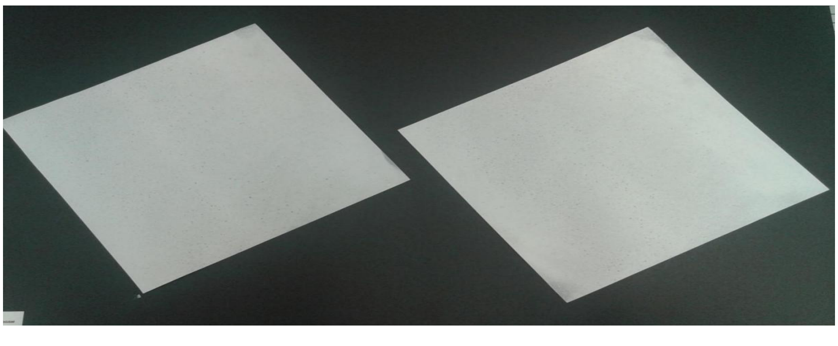

Figure 4. Spray-coated CNFs that are rectangular

The spray-coated NC films in the shapes of circles and rectangles are depicted in **Figures 3** and **4**. The uniform and stable film was created by spray coating. Process variables, like the consistency of the nanocellulose suspension, and engineering variables, like the conveyor's velocity, were used to customize the base weight and thickness of the film. The spray-coated NC film scanning electron microscopy (SEM) micrographs show the cellulose nanofibers' topography, morphology, structure, and distribution of fiber diameters [2,3].

7.1. SEM of spray-coated NC films

Spray-coated NC films display two distinct surfaces, one rough and one smooth, as revealed in the SEM image (**Figure 5**). The film itself was very small and had a smooth, shiny appearance. By applying NC suspension onto a polished metal surface, it was possible to transfer some of the surface roughness to the film, resulting in a smooth and glossy finish. This characteristic facilitates the production of various functional materials such as printed electronics, flexible OLED screens, solar cells, and sensors. The rough side of the film, in contrast to the smooth side, is more porous and has a coarser texture. Various methods were employed to evaluate the surface roughness of the spray-coated nanocellulose films [9].

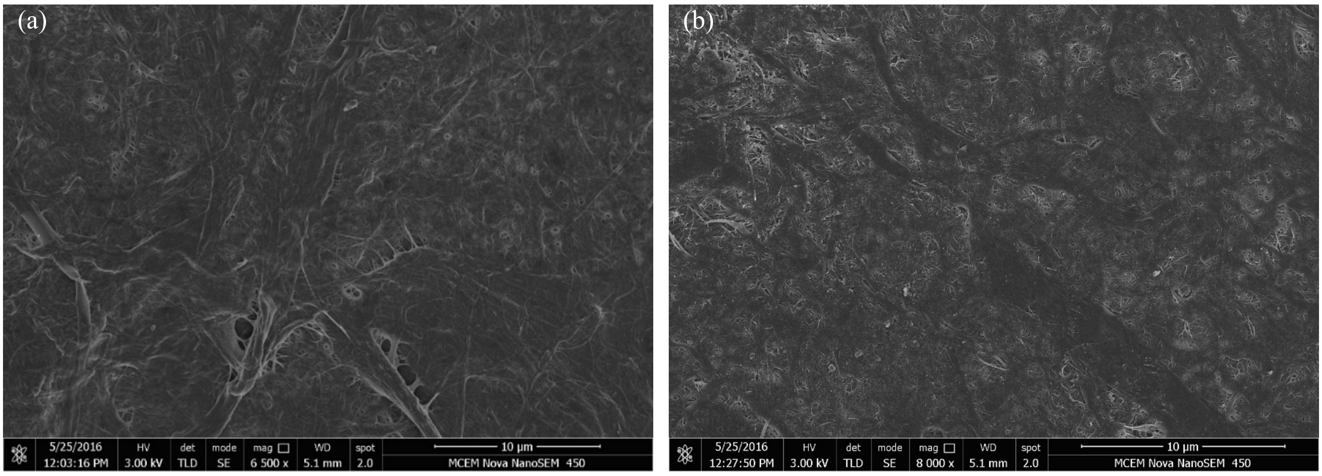

Figure 5. The micrograph of spray coated sheet at 10 μm. **(a)** Rough; **(b)** Smooth

The cross-sectional perspective of spray-coated NC films showed that those sprayed onto the surface were compact and intertwined with neighboring fibers. SEM displayed multiple layers and effective compression of the spray-coated NC films. Additionally, it was indicated that the movement of oxygen, air, and water vapor within the film would take a complex, meandering path. Consequently, the film's ability to act as a barrier is enhanced, potentially transforming it into an impermeable sheet that could serve as a sustainable alternative to synthetic plastics in packaging [10].

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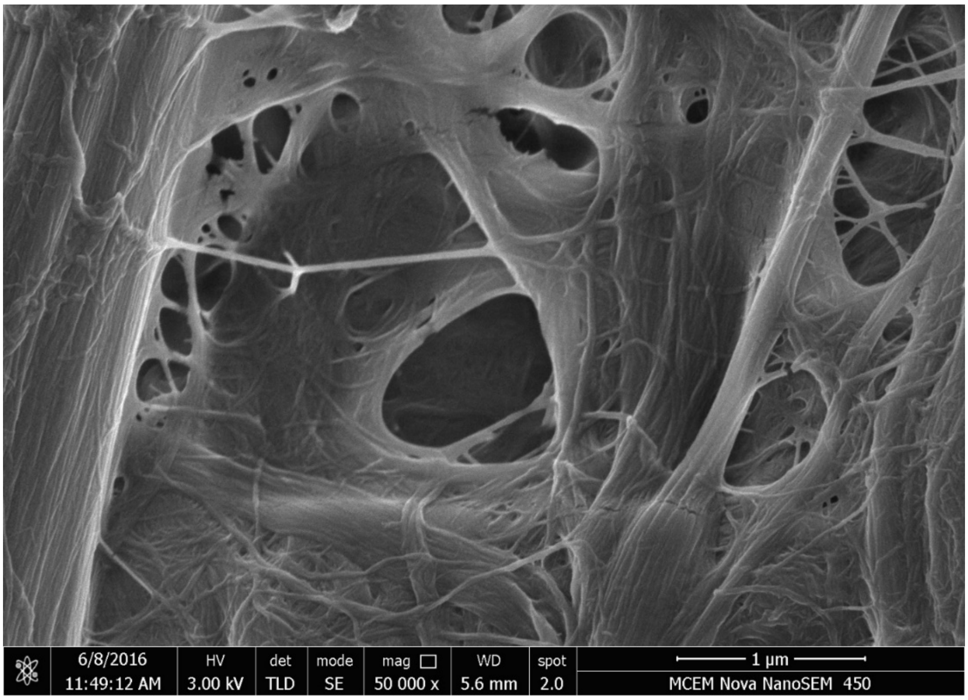

Figure 6. Smooth side of spray-coated CNF

The surface roughness of the film's rough side is comparable to that of the film's surface after vacuum filtration. The rough surface attests to the fact that it is free of pinholes and has dense clumps of cellulose and free fibers of varied sizes. The rough side, or free surface, which is in direct contact with the atmosphere, is depicted in **Figure 5**. Furthermore, the film's surface topography has an asymmetric structure, with the smooth surface having a different pore size than the rough surface. The fibers form a fibrous matrix with varying pore sizes when they are well-connected. Free fibers are pointing toward the atmosphere and possess a high level of surface roughness on the rough side.

7.2. Thickness investigation and thickness mapping of spray-coated CNFs

The most important factor for spray-coated NC films is thickness. Variations in thickness impact the film's tensile and barrier qualities as well as its homogeneity. As previously mentioned, the consistency of the suspension sprayed onto the metal plate can be altered to customize the thickness of the film. Investigating and mapping the thickness of spray-coated nanocellulose films is crucial for ensuring uniformity and consistency in the coating process. There are several thickness mapping techniques available:

(1) Point-by-point mapping: (a) Grid mapping: Measure thickness at various points across the substrate in a grid pattern. This method provides detailed spatial distribution but can be time-consuming; (b) Random sampling: Measure thickness at random locations to get an overall idea of uniformity without exhaustive mapping.

(2) Automated scanning systems: (a) Automated profilometers: Use motorized stages to automatically scan large areas and generate thickness maps; (b) Optical scanners: Employ high-resolution cameras and light sources to create detailed thickness maps quickly.

(3) Mapping and visualization: (a) Software tools: Create 2D and 3D maps to visualize thickness distribution. Software like MATLAB, Python with relevant libraries, or dedicated analysis software from instrument manufacturers can be used; (b) Statistical analysis: (i) Uniformity assessment:

Calculate statistical parameters such as mean, standard deviation, and coefficient of variation to assess uniformity; (ii) Trend analysis: Identify patterns or gradients in thickness distribution to optimize the spraying process.

(4) Applications of uniform thickness nanocellulose films: (a) Quality control in manufacturing: Ensuring consistent coating thickness for industrial applications like protective coatings or electronic devices; (b) Research and development: Investigating the effects of different spraying parameters on film thickness and properties; (c) Biomedical applications: Ensuring uniform coating thickness for nanocellulose films used in wound dressings or biomedical devices.

By employing these techniques and considerations, researchers and manufacturers can accurately investigate and map the thickness of spray-coated nanocellulose films, leading to improved process control and product quality.

The thickness details of the 2.00 wt.% microfibrillated cellulose (MFC)/CNF made by spray coating are displayed in **Figure 7**. **Figure 7** provides the maximum and minimum thickness values for each of the seven films that are depicted in the plot. The plot also includes a reference to the average thickness of each film. The thickness ranges from 200–250 µm. The production of nanocellulose films through spray coating is consistent, with little variation in thickness. A thick and solid basis weight of the film was formed when the 2.00 wt.% was sprayed onto the stainless-steel plate. The movement of the wet film on the plate as a result of handling it causes variations in thickness.

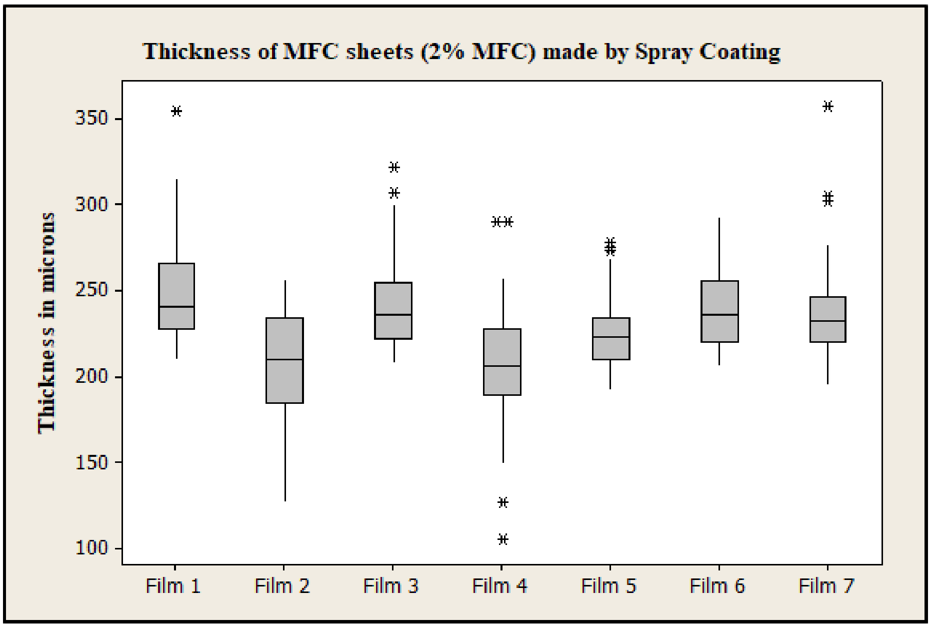

Figure 7. Thickness variation of 2.00 wt.% CNF. * = outlier.

The spray-coated NC film thickness variation of 1.75 wt.%, 1.25 wt.%, and 1.00 wt.% are shown in **Figures 8, 9,** and **10**, respectively. The thickness of the vacuum-filtered NC film is shown in **Figure 11**. The thickness of the films varied the most in these plots, as there is little solid fiber in the 1.25 wt.% and 1.00 wt.% NC suspensions, hence the films behave like watery suspensions. The watery suspension that was sprayed on the plate allowed the suspension to migrate within the wet film, causing a variation in thickness. This can be solved by spraying the NC suspension with a high solid content. The uniformity of the films is improved and thickness variation can be minimized when spraying above 1.5 wt.% NC suspension. There are restrictions on how much NC suspension can be sprayed with the current experimental setup's spray system, which is up to

2.00 wt.%. The viscosity of an NC suspension is reduced upon the addition of carboxymethyl cellulose (CMC) or methylcyclopentadienyl manganese tricarbonyl (MMT) [9,11]. As an alternative, a high-performance spray system was suggested to manage the highly viscous NC suspension to produce CNFs with a high basis weight and thickness [7].

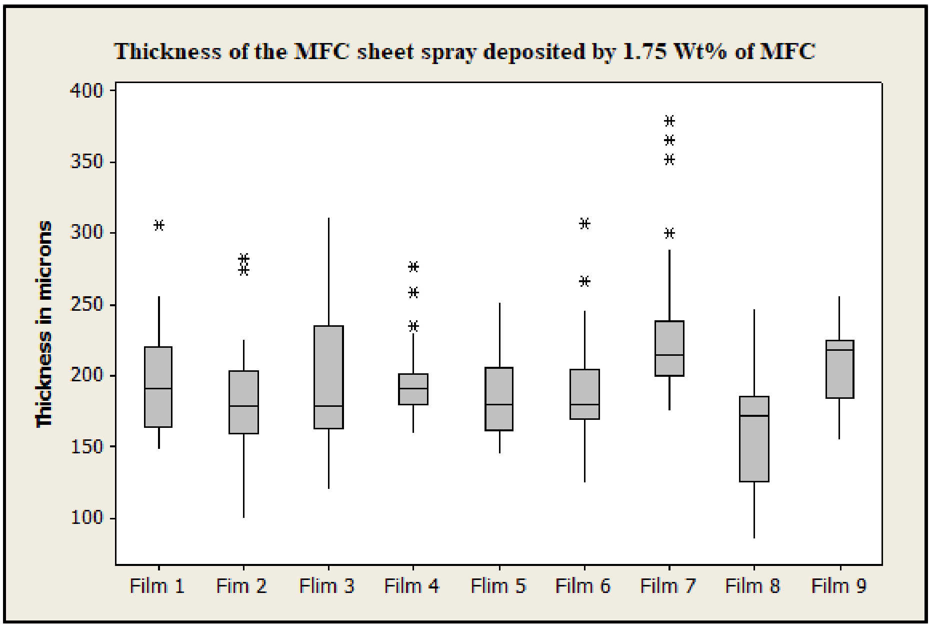

Figure 8. Thickness variation of 1.75 wt.% CNF

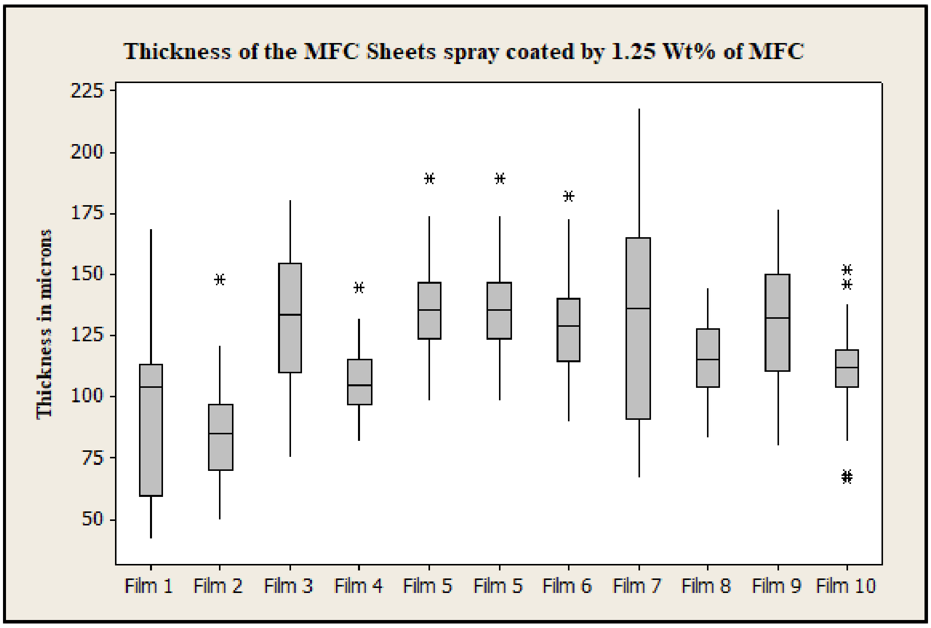

Figure 9. Thickness variation of 1.25 wt.% CNF

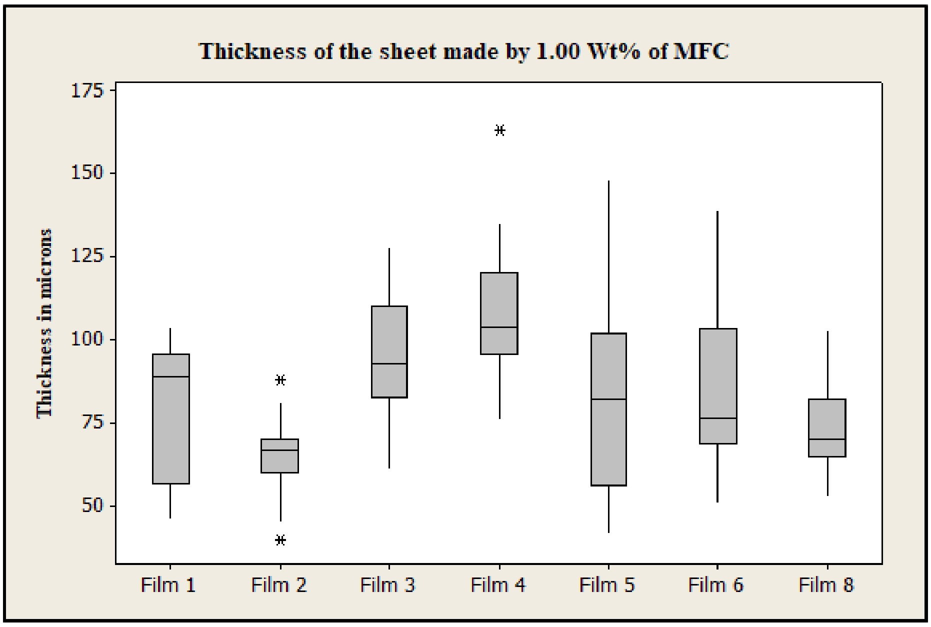

Figure 10. Thickness variation of 1.00 wt.% CNF

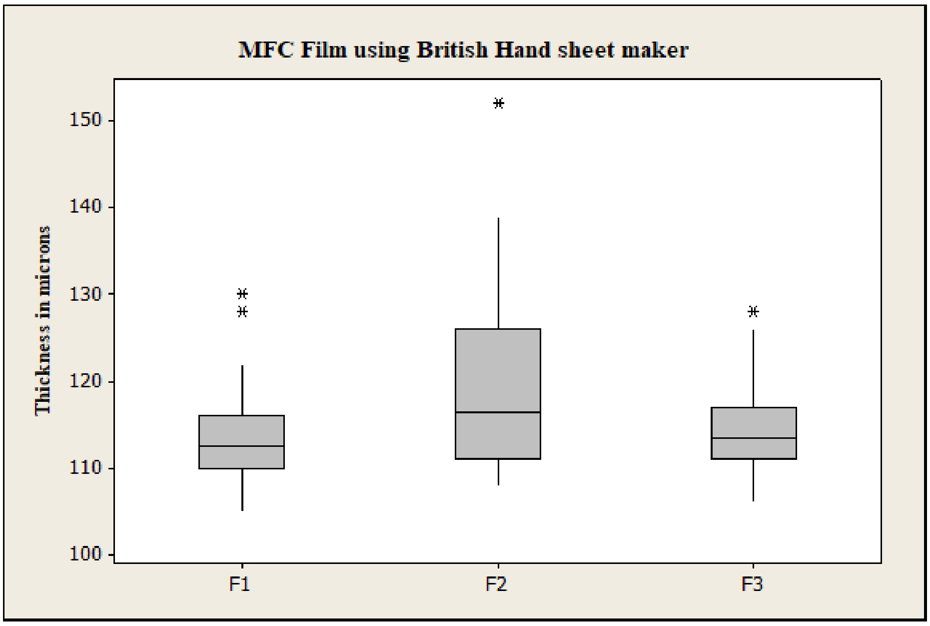

Figure 11. Thickness variation of CNF prepared via vacuum filtration

7.3. Thickness mapping of spray-coated NC films

Using an L&W Thickness Analyzer, the thickness of spray-coated NC films was assessed. To assess the uniformity of the film and compare it with the film obtained through vacuum filtration, thickness mapping was carried out using thickness data. Six regions were identified on the circular section of the film, and these regions' thicknesses were measured. Using Origin Pro 9.1, a contour plot was used to map the thickness of the circular film's central rectangular region. **Figure 12** illustrates the idea of thickness mapping, which is used to assess the film's uniformity [2,12].

Figure 12 illustrates the idea of thickness mapping, which is used to assess the film's uniformity [2,12]. The film's center is where the thickness was measured. Contour plotting is done using the square in the middle of the film. The film's uniformity is confirmed through mapping using the grey point of thickness.

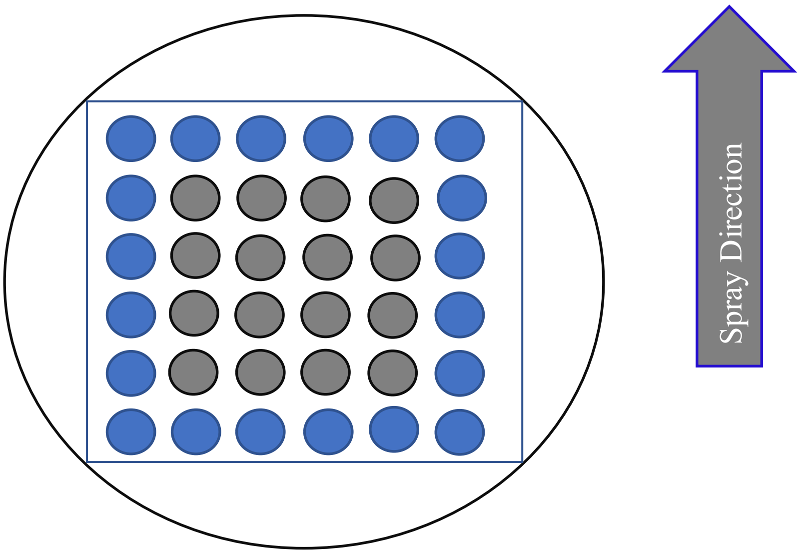

Figure 12. Mapping of the NC film's thickness

The thickness mapping of different spray-coated NC films is displayed in **Figures 13–16**. The thickness mapping verifies the good uniformity and comparability of the spray-coated film with the vacuum-filtered film. The spray-coated film typically has a thicker layer than the vacuum-filtered film. In terms of thickness uniformity, the spray-coated film performs well when compared to vacuum filtration. Maximum variation in film thickness and poor uniformity were observed at the lowest concentration of NC suspension sprayed on the stainless-steel plate. On the steel plate, the low concentration of NC suspension is typically diluted and difficult to remain still. Because of the extreme thickness variation, the film consequently becomes extremely thin and has poor uniformity. Enhancing the suspension consistency during spraying results in a wet film with greater resistance to stagnation and improved thickness and basis weight upon drying. For this reason, the thickness mapping contour plot verifies that the film produced with the lowest concentration of NC had less uniformity than the high-concentration suspension that was sprayed. The uniformity of the spray-coated NC film is comparable to that of the vacuum-filtered film [3].

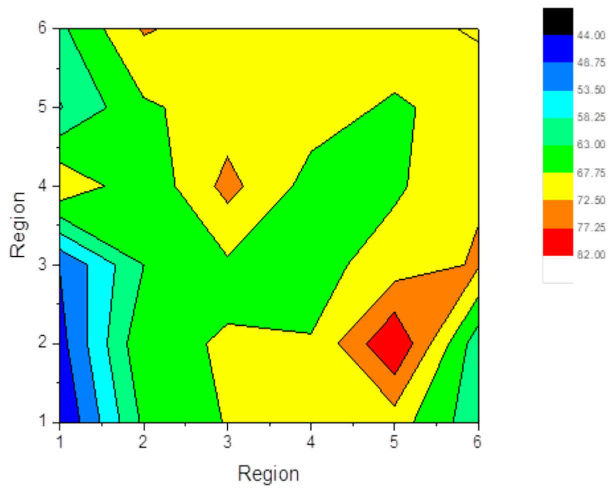

Figure 13. The thickness of mapping 1.00 wt.% spray-coated NC films

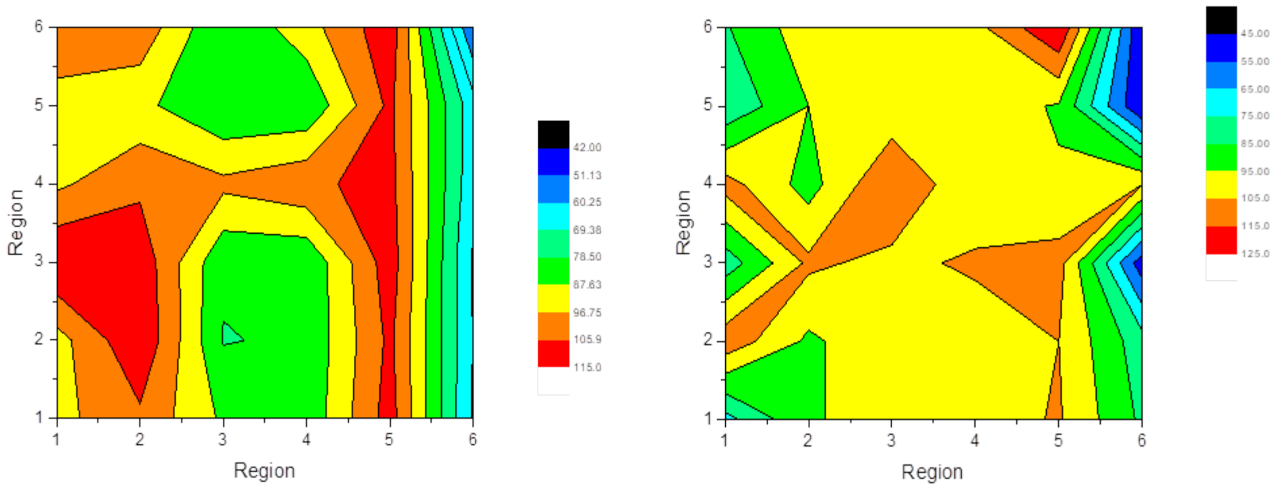

Figure 14. Thickness mapping of 1.25 wt.% of spray-coated NC films

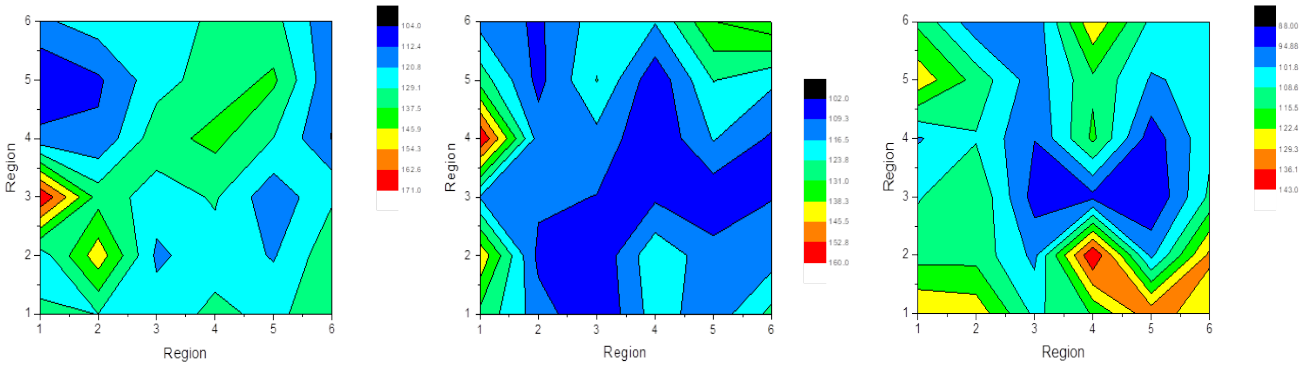

Figure 15. Thickness mapping of 1.5 wt.% of spray-coated NC films

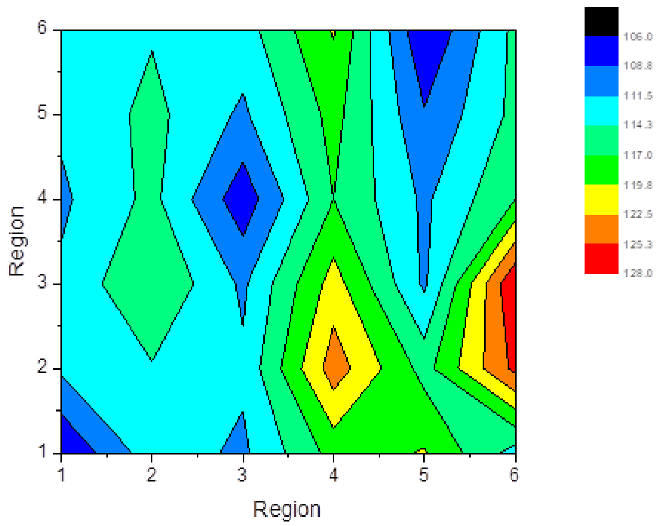

Figure 16. Thickness mapping of NC films prepared via vacuum filtration

7.4. Uniformity of spray-coated NC films

Ensuring the uniformity of spray-coated nanocellulose (NC) films is essential for their effective application and performance. Uniformity pertains to consistent thickness, surface morphology, and material distribution. Here are the key aspects and strategies for achieving and assessing the uniformity of spray-coated nanocellulose films:

(1) Spraying process parameters: (a) Nozzle design and type: (i) Nozzle size: Use a nozzle size that matches the viscosity of the nanocellulose suspension. Smaller nozzles create finer sprays but may clog more easily; (ii) Spray pattern: Select a nozzle that produces a suitable spray pattern (e.g., fan, cone) to cover the substrate evenly; (b) Spray pressure and flow rate: (i) Pressure control: Maintain consistent air pressure. Optimal pressure ensures a stable spray pattern; too high or too low can lead to uneven coatings; (ii) Flow rate: Adjust the flow rate to balance coverage and drying time. A consistent flow rate helps achieve uniform deposition; (c) Distance and angle: (i) Distance to the substrate: Maintain a consistent distance (typically 10–20 cm) between the spray nozzle and the substrate to ensure even distribution; (ii) Spray angle: Keep the spray nozzle perpendicular to the substrate to avoid uneven thickness due to angle variations.

(2) Suspension preparation: (a) Homogeneous dispersion: (i) Mixing: Use mechanical stirring, sonication, or high-shear mixing to ensure the nanocellulose is well-dispersed in the solvent; (ii) Stabilizers: Add dispersing agents or surfactants if necessary to prevent agglomeration and ensure a stable suspension; (b) Concentration: (i) Optimal concentration: Adjust the concentration of the nanocellulose suspension for the desired film thickness and uniformity. Higher concentrations may require more fine-tuning of the spraying parameters.

(3) Environmental conditions: (a) Temperature and humidity: (i) Control environment: Perform the spraying process in a controlled environment where temperature and humidity are regulated to prevent issues like rapid drying or condensation; (b) Airflow: (i) Minimize air currents: Avoid strong air currents that can disrupt the spray pattern and cause uneven deposition.

(4) Substrate preparation: (a) Surface cleanliness: (i) Clean substrate: Thoroughly clean the substrate to remove any contaminants that could affect adhesion and uniformity; (b) Surface activation: (i) Enhance adhesion: Use surface treatments like plasma treatment or chemical etching to improve the adhesion of nanocellulose to the substrate.

(5) Deposition techniques: (a) Multiple passes: (i) Layering: Apply multiple thin layers rather than a single thick layer. This allows for better control over thickness and uniformity; (ii) Overlap: Ensure overlapping spray passes to avoid gaps and streaks; (b) Automated systems: (i) Consistency: Use automated spraying systems with precise control over movement and parameters to ensure consistent coating across large areas.

(6) Thickness and uniformity measurement: (a) Profilometry: (i) Surface profiling: Use contact or non-contact profilometry to measure thickness variations across the coated surface; (b) Microscopy: (i) SEM and AFM: Employ scanning electron microscopy (SEM) or atomic force microscopy (AFM) to examine the surface morphology and detect any non-uniformities; (c) Spectroscopic techniques: (i) Ellipsometry and X-ray reflectivity: Utilize these methods to assess film thickness and uniformity, especially for very thin coatings.

(7) Post-processing: (a) Drying and curing: (i) Controlled drying: Ensure the coated films are dried under controlled conditions to prevent defects like cracking or uneven shrinkage; (ii) Thermal treatment: Use appropriate thermal treatment to improve film adhesion and mechanical properties without causing deformation.

(8) Data analysis and feedback: (a) Statistical analysis: (i) Uniformity metrics: Analyze thickness data statistically to assess the uniformity and identify areas for process improvement; (b) Process optimization: (i) Iterative feedback: Use feedback from uniformity assessments to iteratively optimize spraying parameters and suspension preparation for better consistency.

By carefully managing these factors and continuously monitoring the coating process, it is possible to achieve highly uniform spray-coated nanocellulose films, ensuring their optimal performance in various applications.

7.4.1. Using a paper formation tester to evaluate the uniformity

The paper formation tester, commonly used in the paper industry, can be an effective tool for evaluating the consistency and uniformity of spray-coated nanocellulose films. Here's how it can be applied and the key aspects to consider:

(1) Principle of operation: A paper formation tester typically measures the uniformity of paper sheets by analyzing their optical density variations or other physical properties. This principle can be adapted to assess the consistency of nanocellulose films.

(2) Setup and calibration: (a) Sample preparation: Ensure the nanocellulose film samples are flat and properly sized to fit the tester; (b) Calibration: Calibrate the tester according to the manufacturer's instructions, using standard calibration materials if available.

(3) Testing procedure: (a) Mounting the film: Place the nanocellulose film sample on the test platform, ensuring it is flat and free from wrinkles or bubbles; (b) Measurement settings: Set the tester to the appropriate measurement mode, typically optical density or another relevant metric for film uniformity; (c) Scan and analyze: Run the test to scan the film. The tester will produce a map or profile of the film's properties, highlighting areas of variation.

(4) Data interpretation: (a) Uniformity index: The tester provides a uniformity index or a similar metric that quantifies the consistency of the film; (b) Visual maps: Generate visual maps showing the distribution of optical density or thickness across the film, identifying areas of non-uniformity.

(5) Analyzing consistency and uniformity: (a) Thickness variation: (i) Spot analysis: Use the formation tester to identify spots with significant thickness variations. Compare these results with profilometry or other thickness measurement techniques; (ii) Average thickness: Calculate the average thickness and its standard deviation to assess overall uniformity; (b) Surface defects: (i) Detection of defects: Look for patterns that indicate surface defects such as pinholes, cracks, or aggregates of nanocellulose; (ii) Correlation with other methods: Correlate findings from the formation tester with SEM or AFM images to confirm and detail the nature of the defects; (c) Optical properties: (i) Light transmission/scattering: Analyze how the film transmits or scatters light, providing insight into its optical uniformity, which is crucial for applications like transparent coatings.

(6) Optimizing spray coating process: (a) Parameter adjustments: (i) Spray parameters: Based on the uniformity data, adjust spray parameters (nozzle size, pressure, flow rate) to improve consistency; (ii) Suspension properties: Modify the concentration and dispersion quality of the nanocellulose suspension to enhance film formation; (b) Iterative testing: (i) Process iteration: Perform iterative testing and adjustments. After making changes to the coating process, re-test using the formation tester to evaluate improvements; (ii) Continuous monitoring: Use the formation tester for continuous quality control in a production setting, ensuring each batch of films meets uniformity standards.

Advantages of using a paper formation tester:

(1) Non-destructive testing: The tester allows for non-destructive evaluation of film consistency.

(2) High sensitivity: It can detect subtle variations in thickness and density, providing detailed insights into the uniformity of the film.

(3) Quick and efficient: The testing process is relatively quick, enabling rapid feedback for process optimization.

In conclusion, utilizing a paper formation tester to evaluate the consistency of spray-coated nanocellulose films provides valuable data on their uniformity and quality. This tool was employed to analyze the optical consistency of NC films that were spray-coated and compare them to films produced through vacuum filtration, using paper and paper boards as substrates. By directing light across the film, the device directly measures the film's quality across various length scales ranging from 0.5–60 mm. The Relative Formation Value (RFV) of each component in spray-coated nanocellulose films was determined by comparing the final data with the standard film from vacuum filtration. An RFV value lower than, equal to, or greater than 1 indicates poor, better, or good film uniformity, respectively. By carefully analyzing these results and making informed adjustments to the spraying process, manufacturers can produce films with consistent properties, ensuring their suitability for a wide range of applications.

The optical uniformity of spray-coated NC films, as assessed by the Paper Formation Tester, is illustrated in **Figure 17**, while the standard film created through vacuum filtration is displayed in **Figure 18**. The image indicates that the film exhibits low uniformity, with an RFV value below 1, suggesting the need for more suspension consistency during spraying to form a thick layer of cellulose nanofibers. The presence of air bubbles in the film is a possibility when the NC suspension is sprayed onto the stainless-steel plate, which can affect the film's consistency. The tensile strength, barrier properties, and other bulk characteristics such as thickness and basis weight are closely linked to the uniformity of NC. Increasing the suspension concentration can enhance the film's uniformity. Additionally, the redesign of the spray system and process significantly improves the uniformity of spray-coated NC films [3,5].

Figure 17. Uniformity of spray-coated NC film

Figure 18. Uniformity of vacuum-filtered NC film

7.5. Surface roughness of spray-coated CNFs

A novel technique involves spraying a nanocellulose (NC) suspension onto a polished metal surface to produce smooth NC films. These films have two distinct surfaces: one rough surface facing the air and one smooth surface adhering to the metal. The smoothness of the nanocellulose film is achieved by replicating the smoothness of the base surface, such as a polished stainless steel plate, during the spraying process [2,3]. The development of numerous functional materials, including flexible electronics, printed electronics, solar cells, and microfluidic devices, has been greatly aided by the rough and smooth sides of the film [13]. In these applications, the film's surface roughness is an important factor. Consequently, Parker Surface Print Instrument (PPI), Optical Profilometry (OP), and Atomic Force Microscopy (AFM) were used to measure the film's surface roughness. PPI addresses the films' macroscale roughness, while AFM and OP focus on the films' nanoscale surface roughness.

Surface roughness is a critical parameter in characterizing spray-coated nanocellulose films, as it affects their optical, mechanical, and functional properties. Here's a comprehensive guide on measuring and analyzing the surface roughness of these films:

(1) Importance of surface roughness: (a) Optical properties: Roughness affects light scattering, influencing the transparency and reflectivity of the films; (b) Mechanical properties: Surface roughness can impact the film's adhesion, friction, and wear resistance; (c) Functional performance: For applications like coatings, membranes, or biomedical devices, surface roughness plays a role in wettability, barrier properties, and cell interactions.

(2) Methods for measuring surface roughness: (a) Atomic Force Microscopy (AFM): (i) High resolution: Provides detailed topographical images at the nanoscale; (ii) Quantitative analysis: Measures various roughness parameters, such as Ra (average roughness) and RMS (root mean square) roughness; (b) Profilometry: (i) Contact profilometry: Uses a stylus to physically scan the surface. Suitable for larger areas but might damage delicate films; (ii) Non-contact profilometry: Uses optical methods (e.g., laser) to scan the surface without physical contact; (c) Scanning Electron Microscopy (SEM): (i) Surface imaging: Provides high-resolution images of the surface morphology. Indirectly assesses roughness by analyzing the surface texture; (ii) 3D reconstruction: Advanced SEM techniques can generate 3D surface profiles; (d) Optical microscopy: (i) Interference microscopy: Uses interference patterns to measure surface roughness. Useful for transparent films; (ii) White light interferometry: A non-contact method providing high-resolution 3D surface profiles, suitable for both smooth and rough surfaces.

(3) Surface roughness parameters: (a) Ra (average roughness): The arithmetic average of the absolute values of the surface height deviations measured from the mean plane; (b) Rq (RMS roughness): The root mean square average of height deviations from the mean plane; (c) Rz (maximum height of the profile): The average maximum peak-to-valley height within a sample length; (d) Rt (total height of the profile): The total height difference between the highest peak and the lowest valley.

(4) Optimizing the spray coating process based on roughness data: (a) Spray parameters: Adjust nozzle size, spray pressure, and flow rate to achieve the desired surface roughness; (b) Suspension properties: Optimize the concentration and dispersion of nanocellulose in the suspension to control roughness; (c) Environmental conditions: Control temperature and humidity during the spraying process to minimize roughness variations caused by drying effects.

By systematically measuring and analyzing the surface roughness of spray-coated nanocellulose films, it is possible to optimize the coating process and achieve films with the desired properties for various applications.

8. Film evaluation

8.1. Atomic force microscopy of spray-coated films and films prepared via vacuum filtration

Atomic Force Microscopy (AFM) is a powerful tool for characterizing the surface properties of spray-coated nanocellulose films at the nanoscale. AFM provides high-resolution topographical imaging by scanning a sharp tip across the film surface, allowing it to measure surface roughness, texture, and mechanical properties (see

Figures 19 and **20** as examples). AFM evaluates the film's properties in several ways:

(1) Topographical features: (a) Surface roughness: Quantifies the roughness of the film using parameters such as Ra (average roughness) and RMS (root mean square) roughness; (b) Morphological features: Identifies and measures specific morphological features like fibers, aggregates, or voids.

(2) Uniformity assessment: (a) Thickness uniformity: Analyzes height variations across the scan area to assess the uniformity of the film thickness; (b) Surface defects: Detects and characterizes defects such as pinholes, cracks, or areas of uneven deposition.

Applications of AFM in evaluating nanocellulose films:

(1) Surface quality control: (a) Ensuring uniformity: Uses AFM to ensure that the spray-coated nanocellulose films are uniform in thickness and free from significant defects; (b) Optimizing spraying parameters: Optimizes spraying parameters based on AFM analysis to achieve the desired surface quality and film properties.

(2) Mechanical property analysis: (a) Stiffness and adhesion: Assesses the mechanical properties of the films, crucial for applications requiring specific mechanical performance.

(3) Functional surface characterization: (a) Hydrophilicity/hydrophobicity: Evaluates surface properties that affect wettability, important for applications in coatings, membranes, and biomedical devices.

By utilizing AFM for the detailed characterization of spray-coated nanocellulose films, researchers and manufacturers can gain valuable insights into surface morphology and mechanical properties. This enables the optimization of the coating process for improved film performance.

The AFM micrographs show the surface roughness of vacuum-filtered and spray-coated films. The spray-coated NC film is smoother and less porous compared to the rough side, resembling a polished metal surface. The AFM micrographs reveal that the sprayed film has a glossy, shiny surface, with RMS roughness values of 81.1 nm for the smooth side and 414.0 nm for the rough side in a 10 µm × 10 µm film area. The vacuum-filtered film had RMS roughness values of 417.7 nm for Side 1 and 330.8 nm for Side 2 in a 10 µm × 10 µm inspection area, similar to the rough side of the spray-coated film. **Figures 19** and **20** display the AFM micrographs of spray-coated NC films, while **Figures 21** and **22** show the micrographs of vacuum-filtered films [2].

(a) (b)

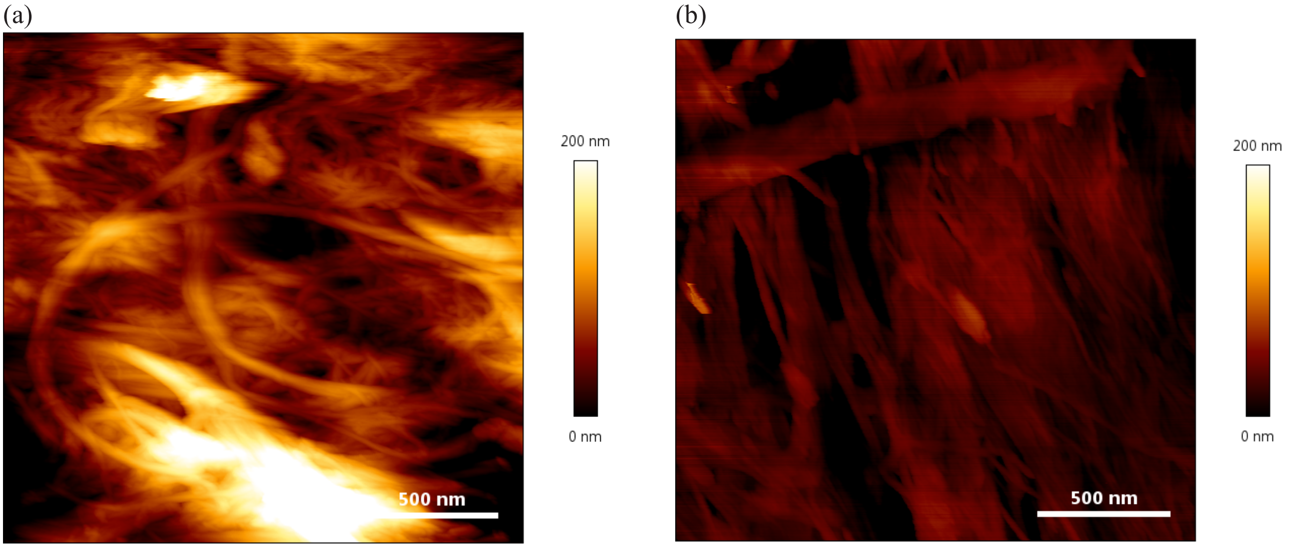

Figure 19. AFM image of a spray-coated sheet prepared by 1.5 wt.% NC spraying on the base surface. **(a)** Free surface; **(b)** Surface exposed to steel

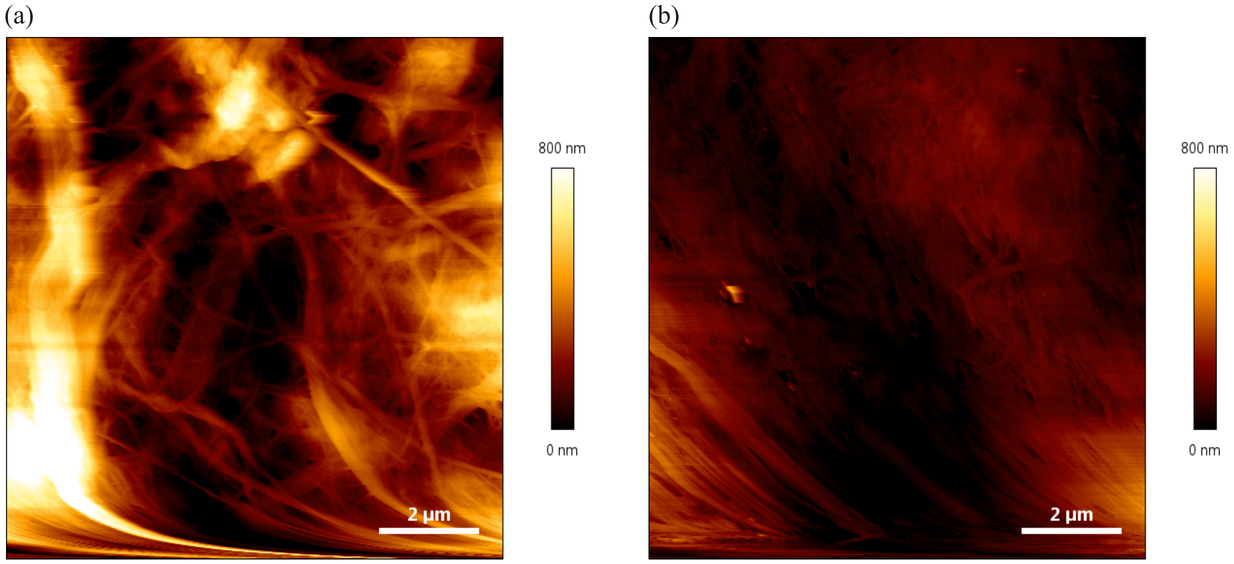

Figure 20. AFM image of spray-coated film. **(a)** Rough; **(b)** Smooth

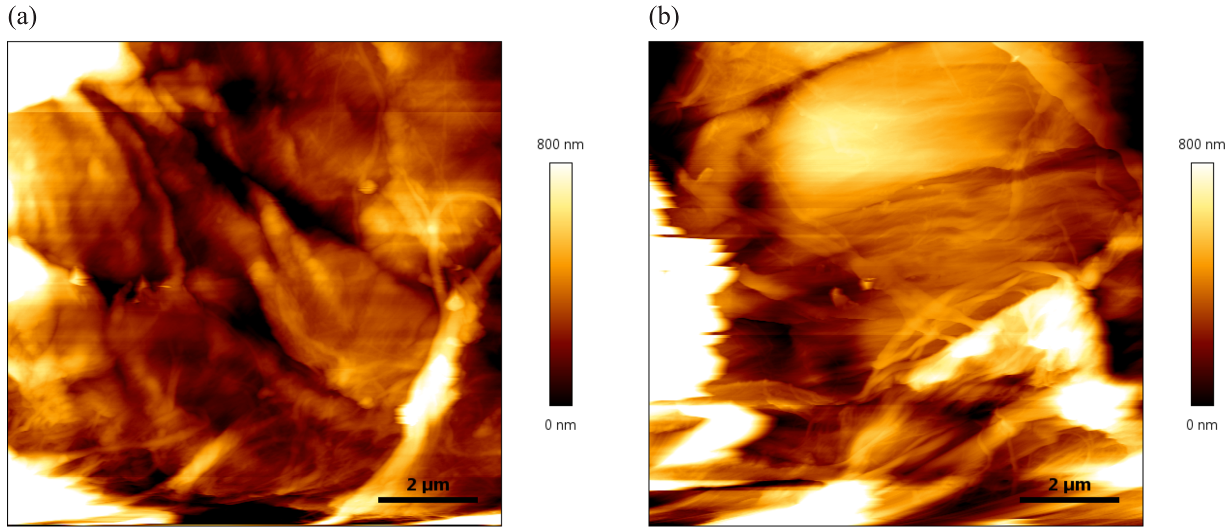

Figure 21. AFM image of the sheet prepared via vacuum filtration. **(a)** Side 1; **(b)** Side 2

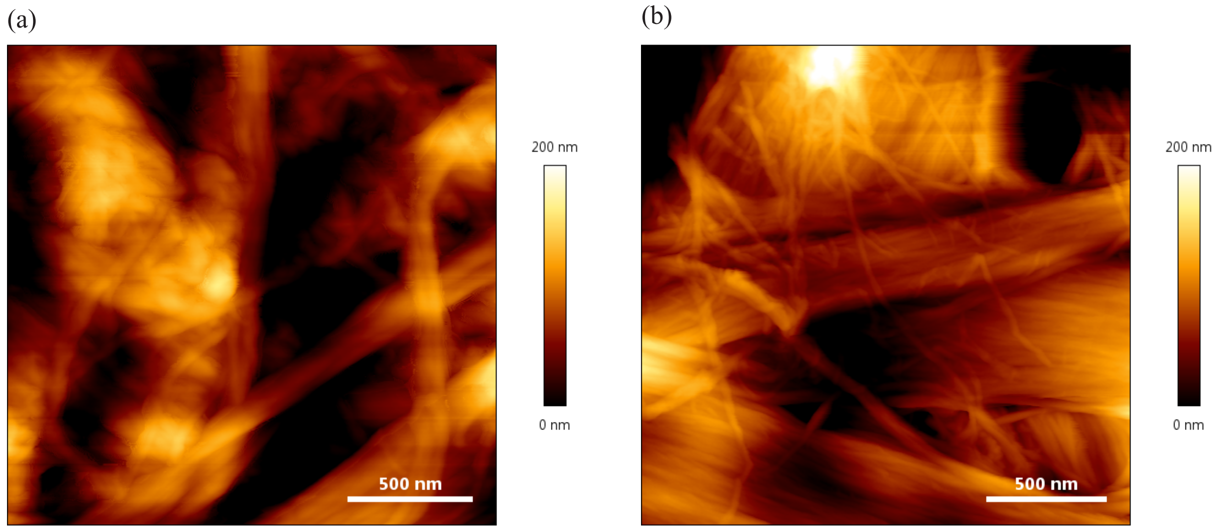

Figure 22. AFM image of the sheet prepared via vacuum filtration. **(a)** Side 1; **(b)** Side 2

8.2. Optical profilometry images for spray-coated NC films and vacuum-filtered NC films

Optical profilometry is an effective non-contact method to measure and visualize the surface topography and roughness of spray-coated nanocellulose films. This technique uses light (often white light or laser) to create detailed 3D maps of a surface by measuring height variations through the analysis of interference patterns or reflected light. Here are key methods for interpreting profilometry images:

(1) Topographical maps: (a) 3D surface maps: Visualize the surface in three dimensions, showing height variations and topographical features; (b) 2D height maps: Display height data in two dimensions with color coding to represent different heights.

(2) Roughness analysis: (a) Surface roughness parameters: Calculate parameters such as Ra (average roughness), Rq (RMS roughness), and Rz (maximum height of the profile) from the height data; (b) Profile sections: Extract cross-sectional profiles to analyze specific features or roughness along a line on the surface.

(3) Feature identification: (a) Defects and anomalies: Identify surface defects such as pinholes, cracks, or aggregates; (b) Uniformity assessment: Assess the uniformity of the coating by examining the consistency of the height data across the scanned area.

Applications of profilometry images:

(1) Quality control: (a) Consistency check: Use profilometry images to ensure consistent film thickness and uniform surface morphology; (b) Defect detection: Identify and quantify surface defects that could affect the film's performance.

(2) Process optimization: (a) Parameter tuning: Adjust spraying parameters based on roughness data to achieve desired surface characteristics; (b) Material analysis: Compare films made with different formulations or processing conditions to optimize the coating process.

By using optical profilometry to analyze spray-coated nanocellulose films, researchers and manufacturers can gain valuable insights into the surface topography and roughness. This allows for fine-tuning the coating process to improve film performance and quality.

Figure 23 shows the optical profilometry images of 5× spray-coated NC films, while **Figure 24** shows the optical profilometry images of 50× spray-coated NC films. The optical profilometry images of 5× and 50× vacuum-filtered NC films were displayed in **Figures 25** and **26**, respectively.

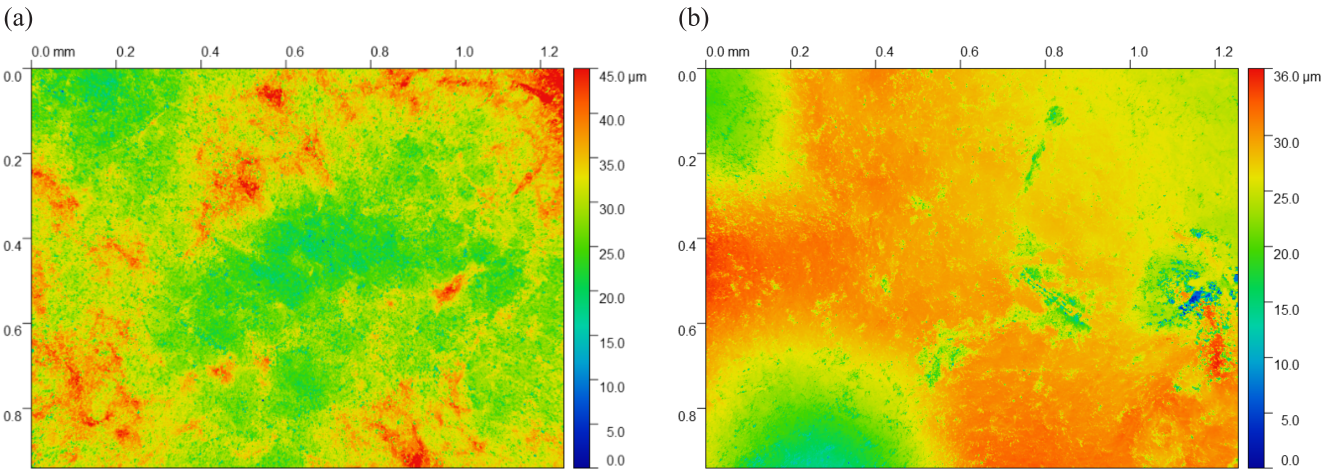

Figure 23. Optical profilometry images of 5× spray-coated NC films. **(a)** Rough side; **(b)** Smooth side

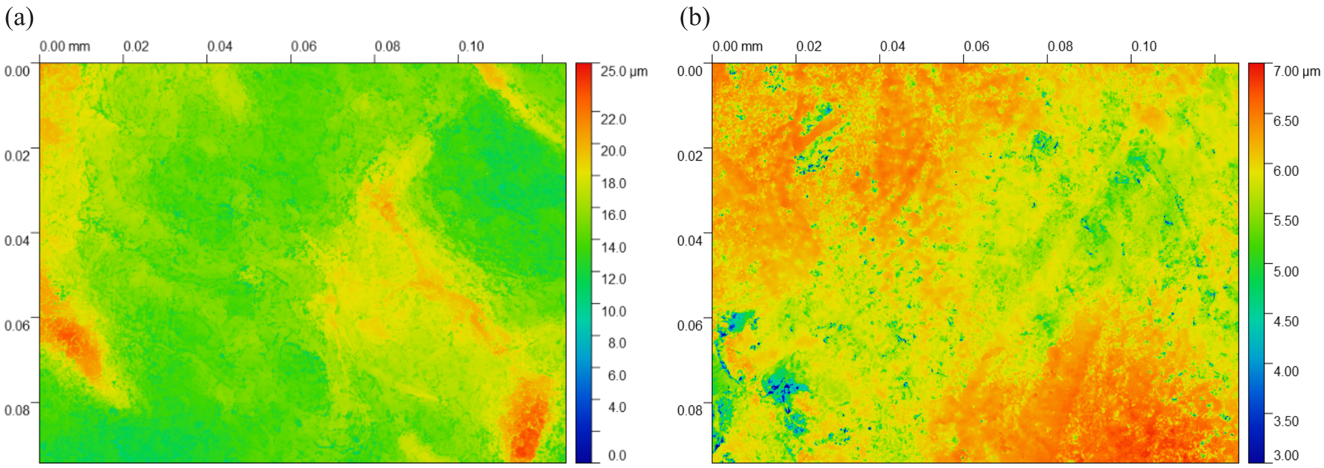

Figure 24. Optical profilometry images of 50× spray-coated NC films. **(a)** Rough side; **(b)** Smooth side

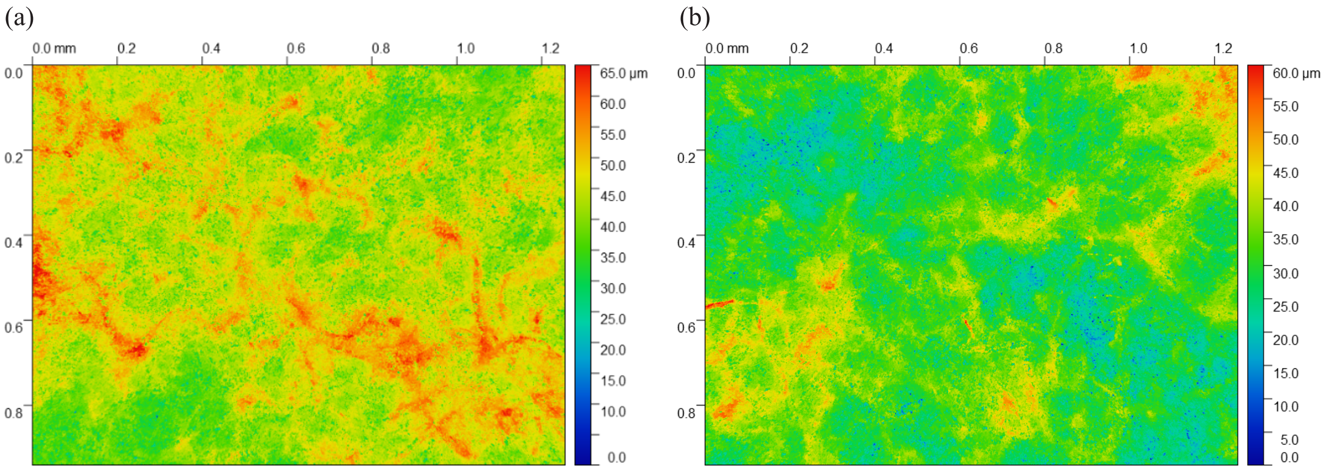

Figure 25. Optical profilometry images of 5× vacuum-filtered NC films. **(a)** Side 1; **(b)** Side 2

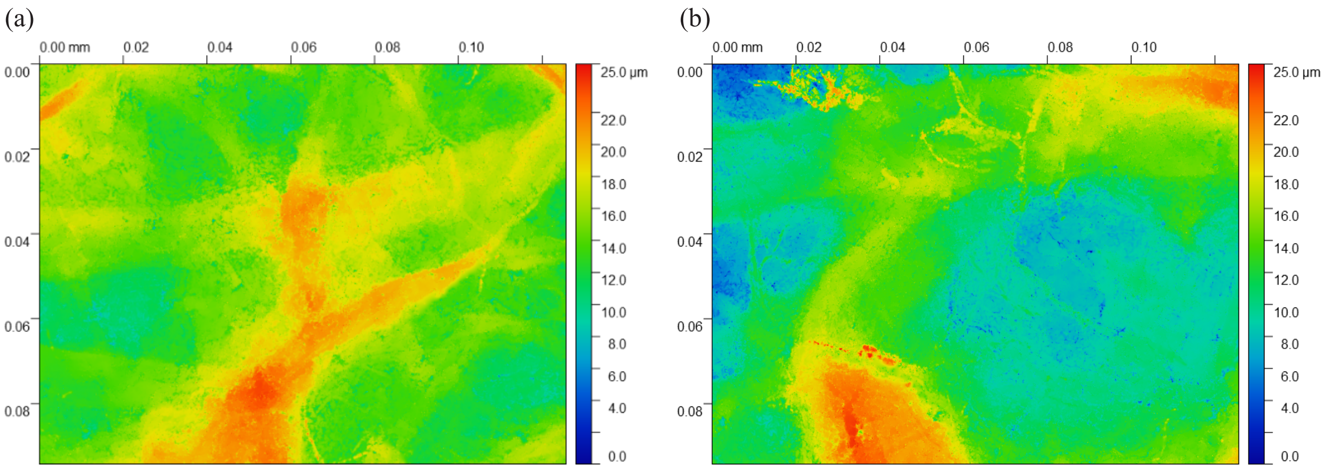

Figure 26. Optical profilometry images of 50× vacuum-filtered NC films. **(a)** Side 1; **(b)** Side 2

8.3. Optical profilometry images of base surface – circular polished stainless-steel plate

Optical profilometry is an effective non-contact method to assess the surface roughness of nanocellulose (NC) films created through spray coating and vacuum filtration. This technique provides detailed surface characterization without causing damage. **Figures 23** to **26** show optical profilometry images of NC films. Comparing NC films produced through vacuum filtration, with RMS roughness values of 2,673 nm on the free side and 3,751 nm on the filter side, the RMS roughness of the spray-coated nanocellulose film measured using optical profilometry at a 1 cm × 1 cm inspection area was 2,087 nm on the rough side and 389 nm on the spray-coated side. These measurements were taken from optical profilometry images with 50× magnification. Additionally, the RMS value for the spray-coated NC film was found to be 5,000 nm on the rough side and 2500 nm on the smooth side based on optical profilometry images with 5× magnification. For the vacuum-filtered NC film, the RMS roughness was 5,900 nm on one side and 6,400 nm on the other. These results suggest that spraying an NC suspension onto a polished metal surface achieves a significant level of smoothness similar to that of a stainless steel plate. The spray method offers a straightforward way to customize the film's surface roughness without additional chemical treatments. Optical profilometry images of a circular polished steel plate are shown in **Figure 27**, replicating this level of surface roughness in the NC films. The RMS roughness of the base surface was measured to be 195 ± 33 nm.

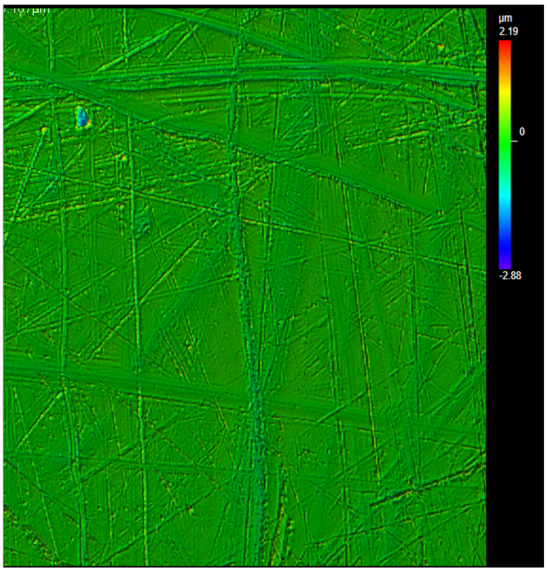

Figure 27. Optical profilometry images of base surface: circular stainless-steel plate (50× magnification)

8.4. Parker surface print instrument for evaluation of macroscale roughness

The surface roughness of paper and paperboard was assessed at the macroscale using the Parker Surface Print Instrument. The surface roughness was 10.8 ± 0.17 µm on the rough side and 5.5 ± 1.4 µm on the smooth side. The NC film that underwent vacuum filtration had a roughness of 10.6 ± 0.3 µm on the filtered side and 9.9 ± 0.1 µm on the free side. These studies conclude that, even at the macroscale, spray-coated films exhibited the lowest surface roughness on the coated side. Another benefit of the spraying method is that it leaves a high-quality finish without the need for additional treatment [3]. **Table 1** summarizes the surface roughness of nanocellulose films as measured by different methodologies.

Table 1. Surface roughness of NC films prepared via spray coating and vacuum filtration

Films preparation method	Technique used	Surface	RMS roughness	
Spray coating	AFM	Rough	414.0 nm (10μm ×10 μm)	51.4 nm (2 μm × 2 μm)
		Smooth	81.1 nm (10μm ×10 μm)	16.7 nm (2 μm × 2 μm)
	Optical profiler 50× image	Rough	2,086.69 nm	
		Smooth	389 nm	
Vacuum filtration	AFM	Free side	417.7 nm (10 μm × 10 μm)	102.3 nm (2 μm × 2 μm)
		Filter side	330.8 (10 μm ×10 μm)	70.7 nm (2 μm × 2 μm)
	Optical profiler 50× image	Free side	2,673 nm (1 cm × 1cm)	
		Filter side	3,751 nm (1 cm × 1 cm)	

9. Bulk properties of NC films

The spray system experimental setup allows for customization of the bulk properties of the NC film by adjusting process and engineering parameters. These bulk properties, including thickness and basis weight, determine the film's tensile characteristics and barrier effectiveness. The relationship between the consistency of the suspension and the resulting film properties is illustrated in **Figure 28**.

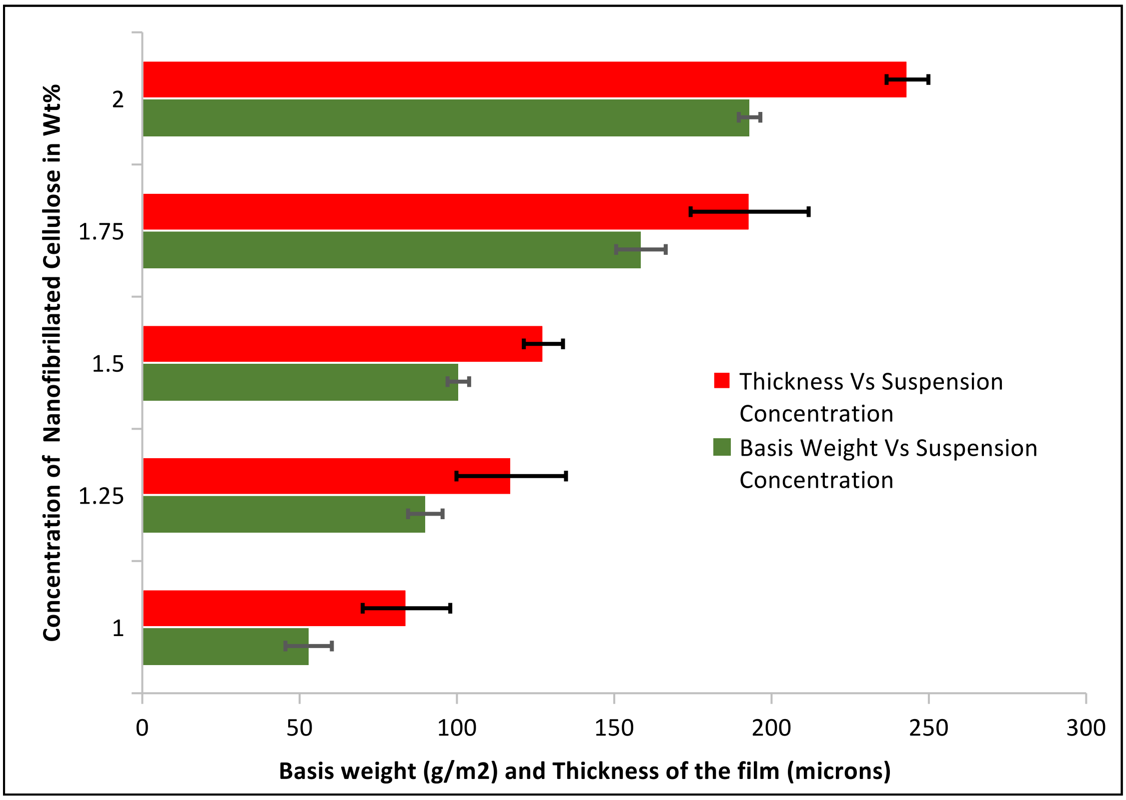

Figure 28. Effect of suspension concentration on the mass per unit area of the NC film using spray coating at a constant velocity of 0.32 cm/sec. Each point is a minimum of 4 replicates, and all replicates were prepared sequentially

The relationship between the consistency of the suspension and the resulting film properties is illustrated in **Figure 28**. **Figure 28** illustrates that when the suspension concentration of NC increased from 1.00 wt% to 2.00 wt%, the mass per unit area of the nano-cellulose film showed a linear increase. The solid content in the suspension primarily regulated the mass of the film per unit area; the conveyor's velocity had little effect on this [2,5]. Additionally, the film produced at the optimal velocity had a high thickness, was free of sheet pinholes, and was packed densely with particles and fibers of different sizes [5]. A maximum of 195 g/m^2 can be achieved by spray coating a nano-cellulose film. The film is homogenous and has a thickness of 240 µm [5]. Professional Wagner spray systems can spray MFC suspensions at concentrations higher than this one. The nanocellulose suspension exhibits time-dependent viscosity and non-Newtonian behavior. The suspension's viscosity is increased by the added fibers and/or particles. Because of this, spraying a suspension with a high solid content can be difficult, and using a nozzle to spray the suspension requires a lot of shear force. Nevertheless, even though the suspension's solid/fiber content is rising, it is challenging for the fibers to break down. As a result, the suspension would lose its fluidity, making it more difficult to handle through a nozzle and increasing the possibility of nozzle clogging. Moreover, the high fiber content of the slurry requires a high shear force to pump and spray [3].

9.1. Mechanical and barrier performance of NC film

Nanocellulose films exhibit unique mechanical and barrier properties, making them suitable for various applications, including packaging, coatings, and biomedical devices. Below is an overview of the key aspects of the mechanical and barrier performance of nanocellulose films, including factors that influence these properties and methods for their evaluation.

(1) Key mechanical properties of nanocellulose films: (a) Tensile strength: The maximum stress the film can withstand while being stretched before breaking; (b) Young's modulus: A measure of the film's stiffness, indicating how much it will deform under stress; (c) Elongation at break: The strain the film experiences before breaking, reflecting its ductility; (d) Toughness: The amount of energy the film can absorb before breaking, combining strength and ductility; (e) Hardness: The film's resistance to surface deformation or indentation.

(2) Factors influencing mechanical properties: (a) Nanocellulose source and type: (i) Cellulose nanofibrils (CNF): Typically provide high strength and flexibility; (ii) Cellulose nanocrystals (CNC): Offer higher stiffness and tensile strength but lower flexibility; (b) Film preparation method: (i) Casting: Produces films with uniform thickness but may have lower mechanical properties due to potential agglomeration of nanocellulose; (ii) Spray coating: Allows control over film thickness and uniformity but may introduce defects affecting mechanical properties; (c) Additives and cross-linking: (i) Plasticizers: Improve flexibility but may reduce strength; (ii) Cross-linking agents: Enhance mechanical strength and stability by forming stronger bonds between nanocellulose fibers; (d) Environmental conditions: (i) Humidity and temperature: Nanocellulose films are sensitive to moisture and temperature, which can affect their mechanical performance.

(3) Testing methods for mechanical properties: (a) Tensile testing: Measures tensile strength, Young's modulus, and elongation at break using a universal testing machine; (b) Nanoindentation: Evaluates hardness and elastic modulus at the nanoscale; (c) Dynamic Mechanical Analysis (DMA): Assesses the viscoelastic properties of the film under varying temperatures and frequencies; (d) Atomic Force Microscopy (AFM): Can be used for nanoscale mechanical testing through force spectroscopy.

(4) Key barrier properties of nanocellulose films: (a) Oxygen permeability: Measures the film's resistance to oxygen transmission, crucial for packaging applications to prevent oxidation; (b) Water vapor permeability (WVP): Assesses the film's resistance to water vapor transmission, important for maintaining product moisture levels; (c) Oil and grease resistance: Evaluates the film's ability to

prevent the passage of oils and greases, relevant for food packaging.

(5) Factors influencing barrier properties: (a) Film density and thickness: Thicker and denser films typically provide better barrier properties; (b) Nanocellulose fiber alignment: Well-aligned nanocellulose fibers can create a more tortuous path for gas and vapor molecules, improving barrier properties; (c) Additives and coatings: Adding hydrophobic or other functional coatings can enhance barrier performance; (d) Environmental conditions: Humidity can swell the nanocellulose, increasing permeability.

(6) Testing methods for barrier properties: (a) Oxygen transmission rate (OTR) test: Measures the rate of oxygen transmission through the film under controlled conditions; (b) Water vapor transmission rate (WVTR) test: Measures the rate of water vapor transmission through the film; (c) Grease resistance test: Evaluates the film's resistance to oil and grease penetration, often using standardized test methods like TAPPI or ASTM.

(7) Applications of nanocellulose films: (a) Packaging: High barrier properties against oxygen and moisture make nanocellulose films ideal for food and pharmaceutical packaging; (b) Coatings: Used as eco-friendly coatings for various surfaces, providing mechanical strength and barrier protection; (c) Biomedical devices: Used in wound dressings and drug delivery systems due to their biocompatibility and mechanical properties.

Nanocellulose films exhibit excellent mechanical and barrier properties, which can be tailored through careful selection of nanocellulose type, additives, and processing methods. By understanding and optimizing these properties, nanocellulose films can be effectively utilized in various high-performance applications. The nanocellulose film coated with a spray has two different surfaces: one that is smooth and glossy, and another that is rough and porous. The quick spraying process enhances the potential of NC films to replace synthetic plastics in barrier applications. Tests showed that the air permeance of the spray-coated nanocellulose film was less than 0.003 μm/Pa/s, indicating its effectiveness in blocking gaseous substances like air. This film has a similar air permeance to the vacuum-filtered film, making it a viable option as a packaging material [3].

One crucial aspect of the NC films is their ability to block water vapor [10,14]. Synthetic plastics typically function as traditional packaging materials and provide an excellent barrier against water vapor [15,16]. One hydrophilic nanomaterial that can prevent water vapor from passing through the film is cellulose nanofiber [16]. Because cellulose nanofibrils are sensitive to water and water vapor in high moisture environments, they get thoroughly wet and loosen the cellulose nanofiber matrix, which widens the pores. Water vapor and other gaseous molecules are consequently transferred more frequently and have greater permeability [20,21]. Spray coating is a useful technique for meeting the criterion of customizing the barrier performance of NC films because it operates independently of the concentration of the NC suspension [3]. As a result, the film's thickness and base weight are easily customized [22]. A dense, shiny film is created by spraying the NC suspension onto a polished metal surface [2]. According to reports, the spray-coated NC film had a water vapor transmission rate of 35.6 ± 8.5 g/m^2 day, while the vacuum-filtered nanocellulose film had a water vapor transmission rate of 33.9 ± 3.5 g/m^2 day [2]. By adding nano clay to the NC matrix, these results can be further decreased and almost equal to the barrier qualities of synthetic plastics [23]. Gaseous and water vapor molecules are transferred across the NC film via a convoluted pathway. The intricate cellulose nanofibrous structures and small pore size improved the barrier performance by expanding the diffusion pathway for gaseous and water vapor [10].

The mechanical characteristics of nanocellulose films play a crucial role in the development of barrier coatings and packaging materials for paper substrates [3,24]. The production methods, types of cellulose fibers, diameter, length, and aspect ratio of the nanofibers all affect the film's tensile properties [7]. Spray-coated NC films' tensile characteristics were similar to those of films produced by vacuum filtration [5]. According to studies, the tensile index of the vacuum-filtered and spray-coated nanocellulose films were 62.3 ± 3.4 Nm/g and 60.2 ± 1.5 Nm/g, respectively [2]. It was discovered that the vacuum-filtered film had a tensile stiffness of 941 ± 78 kN/M, while the spray-coated film had a tensile stiffness of 946 ± 86 kN/M [2]. These results imply that the strength of the spray-coated NC films was equivalent to that of the vacuum-filtered film. By forming hydrogen bonds with nearby fibers, the cellulose nanofibers in the NC films create compactness, which leads to good strength and modulus. The film's thickness and base weight have an impact on its strength as well. Spraying can only be used to modify the film's thickness and base weight by changing the concentration of the NC suspension and other engineering parameters within the spray coating test system. Thus, efficient spraying can be used to engineer the strength of NC films [5].

10. Critical parameters in spray coating

To quickly produce nanocellulose films, a suspension of nanocellulose (NC) is sprayed onto a smooth metal surface. It takes less than a minute for the wet film to form on the plates during this process. In a study, it took about 50.2 seconds to produce a 15.9 cm diameter film. Creating a film with the same diameter and a base weight of 60 g/m^2 required approximately 10 minutes. To reduce production time, one can increase the suspension consistency during vacuum filtration, which leads to longer dewatering times. However, the duration of the spraying process is not influenced by the suspension consistency. Several important factors play a role in influencing the creation of NC films through spraying [7].

Fabricating free-standing nanocellulose films via the spraying process involves several critical parameters that must be controlled to ensure the desired film properties and quality. Here's a comprehensive overview of these parameters:

(1) Nanocellulose suspension characteristics: (a) Concentration: The concentration of nanocellulose in the suspension affects the viscosity and the final thickness of the film. Optimal concentration ensures uniform deposition and film formation; (b) Viscosity: The viscosity of the suspension influences the sprayability and uniformity of the film. It should be low enough to allow smooth spraying but high enough to avoid excessive spreading; (c) Stability: The suspension should be stable without significant aggregation or sedimentation of nanocellulose particles to ensure consistent spraying and film quality; (d) Additives: Additives such as surfactants, dispersants, or plasticizers may be used to enhance suspension stability, film flexibility, or other desired properties.

(2) Spraying equipment and conditions: (a) Nozzle type and size: The type and size of the nozzle determine the droplet size and spray pattern. Fine nozzles produce smaller droplets and more uniform films; (b) Spray pressure: The pressure at which the suspension is sprayed affects the velocity and distribution of droplets. Optimal pressure ensures uniform coverage without splattering; (c) Spray distance: The distance between the nozzle and the substrate affects the drying rate and uniformity of the film. Too close can cause uneven deposition, while too far can result in excessive evaporation before deposition; (d) Spray angle: The angle of spraying influences the overlap of spray passes and the uniformity of the film. Consistent angles ensure even coverage.

(3) Substrate preparation and properties: (a) Surface energy: The surface energy of the substrate affects the wettability and adhesion of the nanocellulose suspension. Pre-treatments (e.g., plasma treatment) can enhance wettability; (b) Temperature: The temperature of the substrate can influence the drying rate of the deposited suspension. Warm substrates can accelerate drying, reducing the risk of run-off; (c) Texture and roughness: The physical texture and roughness of the substrate affect the mechanical

interlocking and adhesion of the film.

(4) Environmental conditions: (a) Humidity: Ambient humidity affects the drying rate and can influence the film's final properties. Controlled humidity conditions can prevent defects such as cracking or uneven drying; (b) Temperature: Ambient temperature also impacts the drying rate and film formation. Consistent temperatures help in achieving uniform films.

(5) Spraying technique: (a) Spray pattern: The pattern in which the suspension is sprayed (e.g., zigzag, circular) affects the uniformity of the film. Overlapping spray passes can ensure complete and even coverage; (b) Spray speed: The speed of the nozzle movement influences the thickness and uniformity of the film. Consistent speed ensures even deposition; (c) Layering: Spraying in multiple thin layers rather than a single thick layer can improve film uniformity and reduce the risk of defects.

(6) Drying and post-treatment: (a) Drying method: The method used to dry the sprayed film (e.g., air drying, oven drying) affects the film's mechanical properties and morphology. Controlled drying conditions can prevent issues like warping or shrinkage; (b) Post-treatment: Post-treatments such as annealing or chemical cross-linking can enhance the film's properties, including mechanical strength and stability.

Key considerations for free-standing nanocellulose films:

(1) Thickness control: Achieving the desired film thickness requires precise control over the spraying parameters and the concentration of the nanocellulose suspension.

(2) Uniformity: Uniform deposition is critical for the mechanical integrity and performance of the film. Fine-tuning the spraying parameters and technique is essential for uniform films.

(3) Adhesion and release: Ensuring good adhesion during the spraying process and easy release of the free-standing film from the substrate can be challenging. Substrate treatments or the use of sacrificial layers can aid in this process.

(4) Mechanical properties: The mechanical properties of the final film, such as tensile strength and flexibility, are influenced by the nanocellulose type, suspension formulation, and spraying conditions.

Fabricating free-standing nanocellulose films via spraying involves careful consideration and control of multiple parameters, from the characteristics of the nanocellulose suspension to the environmental conditions during spraying. By optimizing these parameters, high-quality, uniform, and mechanically robust nanocellulose films can be produced, suitable for various advanced applications.

10.1. NC suspension consistency

One of the most important factors in the spraying process is suspension consistency [2]. The thickness and base weight of the film increases linearly with the suspension consistency [2,12]. The suspension cannot be sprayed with the current spray system at a consistency of more than 2.00 wt.% [5]. Rheology modifiers, such as CMC and MMT nano clay can be added to address this issue [9,11]. To create the composite films, the suspension can therefore be sprayed at a higher solid concentration. The flow of the spray jet from the spray nozzle was another observation regarding the spraying nanocellulose suspension. The low viscosity of the suspension causes the spray jet to become wide and erratic at low suspension concentrations (0.25 wt%–0.5 wt%). When the spray jet strikes the base surface, it is reflective [18]. This leads to poor uniformity [5]. This situation may be appropriate for applying barrier layers of NC to paper substrates [18,25]. Water in the NC suspension was absorbed by the paper substrates, which resulted in a coating of cellulose nanofibers on the paper surface and reduced spray jet reflection. A higher solid concentration of cellulose nanofibers reduced the reflection of the spray jet from the base surface when spraying at high suspension concentrations (0.75 wt%–2.25 wt%). The spray jet's width and velocity were both lower at higher concentrations due to the suspension's increased viscosity. This resulted in better uniformity. Generally, thick and dense freestanding NC films can be made by spraying a high concentration of nanocellulose suspension [5].

10.2. Adhesion between NC suspension and base surface

The formation of a film or barrier layer on substrates is largely dependent on the wettability and adhesion of the nanocellulose (NC) suspension on the base surface. The circular stainless steel plate was normally highly polished and had a hydrophobic surface. When low concentrations of solid NC suspension are sprayed onto the plates, it creates liquid droplets that are very mobile and hinder the formation of films. Visual observation revealed a significant contact angle between the sprayed NC droplets and the circular stainless steel plate. To address this, the plate was pre-wetted by spraying a high-concentration NC suspension on it. However, the spray-coated suspension remained highly stagnant [5]. On the other hand, spraying low concentrations of NC suspension on paper substrates makes it easy to wet the paper surface and effectively forms a barrier coating on the paper surface [25]. Water from the spray-coated NC suspension is instantly absorbed by the paper substrates [3], enhancing the coating's wettability and adhesion [7].

Adhesion between nanocellulose suspensions and base surfaces is a critical aspect in various applications, including coatings, adhesives, and composites. The effectiveness of this adhesion depends on several factors, including the properties of the nanocellulose, the nature of the base surface, and the interaction between them. Here's a detailed overview of these factors and their influence on adhesion as well as methods to enhance adhesion and key considerations:

(1) Factors influencing adhesion: (a) Properties of Nanocellulose: (i) Type of nanocellulose: Nanocellulose can exist as cellulose nanocrystals (CNCs), cellulose nanofibers (CNFs), or bacterial nanocellulose (BNC). Each type has distinct properties affecting adhesion. CNCs have high crystallinity and aspect ratio, CNFs have a network structure with high entanglement, and BNC has a three-dimensional network structure; (ii) Surface chemistry: The presence of hydroxyl groups on nanocellulose surfaces allows for hydrogen bonding with the base surface. Chemical modifications (e.g., carboxylation, phosphorylation) can enhance or tailor these interactions; (iii) Particle size and shape: Smaller particles with higher surface area can improve adhesion by providing more contact points. (b) Properties of the base surface: (i) Material composition: Different materials (e.g., metals, polymers, glass) have varying surface energies and chemical functionalities, affecting how well nanocellulose adheres; (ii) Surface roughness: Rough surfaces can enhance mechanical interlocking, improving adhesion. However, very rough surfaces might lead to uneven coating; (iii) Surface treatment: Pretreatments like plasma treatment, chemical etching, or coating with adhesion promoters can enhance surface energy and functional groups, improving adhesion; (c) Interactions between nanocellulose and base surface: (i) Hydrogen bonding: Nanocellulose can form hydrogen bonds with polar surfaces, significantly enhancing adhesion; (ii) Van der Waals forces: These forces contribute to adhesion but are generally weaker than hydrogen bonding; (iii) Covalent bonding: Through chemical modification, covalent bonds can be formed between nanocellulose and the base surface, providing strong adhesion; (iv) Electrostatic interactions: Charged nanocellulose particles can adhere to oppositely charged surfaces.

(2) Methods to enhance adhesion: (a) Surface modification of nanocellulose: (i) Functionalization: Introducing functional groups such as carboxyl, amine, or epoxy groups can improve compatibility with the base surface; (ii) Polymer coating: Coating nanocellulose with polymers can enhance adhesion by providing additional functional groups or improving mechanical properties; (b) Surface Treatment

of the base surface: (i) Plasma treatment: Plasma can introduce functional groups and increase surface energy, enhancing adhesion; (ii) Chemical etching: Creates micro-scale roughness, enhancing mechanical interlocking; (iii) Adhesion promoters: Applying primers or coupling agents (e.g., silanes) can improve bonding between nanocellulose and the base surface; (c) Composite formation: (i) Hybrid materials: Combining nanocellulose with other materials (e.g., polymers, nanoparticles) can improve adhesion by leveraging the strengths of multiple components; (ii) Layer-by-layer assembly: This technique can build up alternating layers of nanocellulose and complementary materials to enhance overall adhesion.

(3) Key considerations: (a) Achieving uniform adhesion across large surfaces can be challenging. Research into optimizing processing conditions is ongoing; (b) Scaling up the production and application processes for industrial use is a key area of focus; (c) Ensuring that adhesion properties are maintained under various environmental conditions (e.g., humidity, temperature) is important for practical applications.

The adhesion between nanocellulose suspensions and base surfaces is influenced by a complex interplay of factors related to both the nanocellulose and the base surface. By understanding and optimizing these factors, nanocellulose can be effectively used in a wide range of applications, from coatings and adhesives to advanced composites. Ongoing research and development will continue to enhance these adhesion properties, making nanocellulose an increasingly valuable material in various fields.

10.3. Spray distance in the experimental setup

The spray pattern, spray width, and spray jet from the spray nozzle are all influenced by the spray distance—the distance between the spray tip and the base surface. To develop uniform film formation and coating on the substrates, an ideal distance must be maintained. The consistency of the NC suspension determines the optimal spray distance. If the suspension's viscosity is low, a good distance—roughly 50 cm—should be maintained to minimize the spray jet's reflectivity from the base surface and promote good film formation. When there is high suspension consistency, the distance should be kept small, about 30 cm. This is so that high-suspension NC deposits on the plate with ease and forms the film efficiently without reflecting from the stainless steel plate [5,25].

10.4. Base surface

The surface morphology and topography of the base surface determine the formation of a barrier layer or film [9]. For films to form, the base surface's wettability is crucial. Permeable and impermeable substrates are two categories of base surfaces. To improve the barrier performance, breathable substrates like newsprint paper, packaging paper, and blotting paper are used as the base surface for coating NC [26]. Water readily wets blotting paper and newsprint paper, and the adhesion force between the paper surface and the NC coating is easily regulated. Barrier coating on the paper substrates requires a low concentration of NC suspension. Silicon wafers, regular stainless steel plates, super polished stainless steel plates, and circular stainless steel plates are impermeable substrates. As stated above, these substrates' smoothness increases in the same order. The spray-coated NC suspension then forms droplets with a high contact angle and poor wettability because the surface is hydrophobic. NC with high-suspension consistency nanocellulose can be used to solve this [9].

10.5. Spray systems

The professional Wagner spray system, normally used in paint spray practice, is utilized in this study. By improving the spray system, spraying can be performed better. The lowest spray system version cannot spray more than 2.00 wt.% of a high suspension concentration. This is because the spray pattern and spray jet formation are blocked by the accumulation of cellulose nanofibers in the spray nozzle. The apparatus was able to manage the high suspension nanocellulose concentration despite an increase in the spray system version without any blockages of fiber aggregates in the spray nozzle. The suspension of NC exhibits non-Newtonian behavior and a time-dependent viscosity. The suspension's viscosity is increased by the added fibers and/or particles. Because of this, spraying a suspension with a high solid content can be challenging, and using a nozzle to spray the suspension requires a lot of shear force. The disintegration of the fibers in the suspension is challenging, despite the suspension's increased solid/fiber content. As a result, the suspension would lose its fluidity, making it more difficult to handle through a nozzle and increasing the possibility of nozzle clogging. Moreover, the high fiber content of the slurry requires a high shear force to pump and spray [7].

10.6. Velocity of the conveyor

Another crucial factor in the experimental spray system setup is the conveyor's velocity [2]. The thickness and base weight of the desired film to be produced are produced by variations in the conveyor's velocity, which has a base surface for coating, once the ideal NC concentration with a good solid content has been determined and fixed for spraying on it. The NC suspension was more heavily deposited on the base surface when the conveyor was moving at a low speed, which produced a film with good uniformity and thickness. Thin film formation occurs when the conveyor travels at a higher speed as less NC is deposited by spraying. The task at hand involves operating the conveyor in the experimental spray system at the lowest possible velocity or speed. The production of high-quality, uniform NC film depends on the proper matching of the conveyor's velocity with the suspension consistency [5].

10.7. Other engineering parameters for improving the spray system

A wet layer of NC is created when the suspension is sprayed onto the polished metal surface. The rapid process of removing water from wet spray-coated NC film is necessary for drying. Certain waste heat from the paper and pulp industries can be used to speed up the drying process. The production of NC film and continuous drying using an infrared (IR) dryer and roll-to-roll coating integrated with a spray system can be used in continuous processes. To further enhance the spraying procedures and attain the highest level of film uniformity, computers can be used to simulate spray patterns and conduct jet analysis [7].

11. Biomedical application of spray-coated free-standing NC films

Spray-coated free-standing nanocellulose (NC) films have several promising biomedical applications due to their unique properties and versatility. NC films can be spray-coated onto wounds to create a thin, protective layer that promotes wound healing. These films possess high water-holding capacity, excellent biocompatibility, and the ability to maintain a moist environment, which is beneficial for wound healing. They can also be loaded with therapeutic agents, such as growth factors or antimicrobial agents, for controlled release to enhance the healing process. Additionally, NC films can be used as drug delivery systems. By incorporating therapeutic agents into the film or coating the film with a drug-loaded layer, these films can provide sustained release of drugs, allowing for localized and controlled delivery to specific target sites. The porous nature of NC films facilitates drug loading and release kinetics, making them suitable for applications in tissue engineering and regenerative medicine.

Spray-coated NC films can serve as scaffolds for cell growth and tissue engineering. The films provide a supportive structure with a high surface area-to-volume ratio for cell attachment and proliferation. They can be tailored to mimic the extracellular matrix, allowing for cellular interaction, nutrient diffusion, and the regeneration of various tissues, such as bone, cartilage, and skin.

Nanocellulose films spray-coated onto medical devices, such as implants or stents, can provide bioactive coatings that enhance biocompatibility and reduce the risk of adverse reactions. These coatings can promote cell adhesion and prevent bacterial adhesion, improving the performance and safety of the devices. Moreover, the films can be functionalized with bioactive molecules or growth factors to further enhance tissue integration and regeneration. Additionally, they can be utilized in biosensors and diagnostic devices. The films can serve as a matrix for immobilizing biomolecules, such as enzymes or antibodies, that selectively interact with target analytes. This enables the development of sensitive and specific detection platforms for applications ranging from healthcare diagnostics to environmental monitoring.

Free-standing NC films have emerged as a promising platform for various biomedical applications due to their unique combination of properties. Derived from natural cellulose, they are well-tolerated by the human body. They possess remarkable strength and flexibility, enabling them to withstand biological stresses. Their high porosity allows for efficient cell interaction and nutrient diffusion. They offer ample space for the immobilization of biomolecules like proteins, drugs, and enzymes, and their surface chemistry and functionalities can be easily modified for specific applications.

Tissue engineering holds great promise in regenerative medicine, and the application of nanocellulose films in this field has garnered significant interest. Nanocellulose, derived from natural sources like wood or plants, offers unique properties such as biocompatibility, biodegradability, and mechanical strength, making it an ideal candidate for tissue engineering applications. Nanocellulose films can serve as scaffolds for cell growth and tissue regeneration due to their porous structure, which allows for nutrient and oxygen exchange. These films can mimic the extracellular matrix, providing support and cues for cell adhesion, proliferation, and differentiation. Moreover, the nanoscale structure of cellulose promotes cell-cell interactions and tissue organization. Incorporating bioactive molecules or growth factors into nanocellulose films can further enhance their functionality by promoting specific cell behaviors or tissue formation. Additionally, the mechanical properties of nanocellulose films can be tailored to match those of various tissues, providing support during the healing process. Overall, the use of nanocellulose films in tissue engineering shows great potential for developing advanced regenerative therapies and medical devices that can address various tissue repair and replacement needs.

Nanocellulose films have shown great potential in drug delivery applications due to their unique properties. These films can be used as carriers for drug molecules, providing a controlled release over time. The high surface area and porosity of nanocellulose films allow for efficient loading of drug molecules, while their biocompatibility and biodegradability make them suitable for use in medical applications. Additionally, nanocellulose films can be easily modified to tailor their drug delivery properties, such as release kinetics and targeting capabilities. By incorporating nanocellulose films into drug delivery systems, researchers are exploring the possibility of improving drug efficacy, reducing side effects, and enhancing patient compliance. Furthermore, the renewable and sustainable nature of nanocellulose makes it an attractive material for pharmaceutical applications. Overall, the drug delivery application of nanocellulose films holds great promise in revolutionizing the field of medicine by offering innovative solutions for targeted and controlled drug delivery.

Nanocellulose films have garnered significant attention in the field of tissue engineering due to their exceptional mechanical properties, biocompatibility, and versatile surface chemistry. Here’s a detailed overview of their applications in tissue engineering:

(1) Scaffold for tissue regeneration: (a) Skin tissue engineering: Nanocellulose films can be used as wound

dressings to promote healing by providing a moist environment and supporting the growth of skin cells. They can be infused with antimicrobial agents to prevent infections; (b) Bone tissue engineering: Due to their high mechanical strength and bioactivity, nanocellulose films are used to create scaffolds that support bone regeneration. They can be combined with hydroxyapatite to enhance osteoconductivity; (c) Cartilage tissue engineering: The flexibility and biocompatibility of nanocellulose films make them suitable for cartilage repair. They provide a conducive environment for chondrocytes to regenerate cartilage tissue.

(2) Drug delivery systems: (a) Localized drug delivery: Films can be designed to release growth factors, antibiotics, or anti-inflammatory drugs directly at the injury site, enhancing tissue regeneration and reducing systemic side effects; (b) Sustained release: The nanocellulose matrix can be tailored to provide a sustained release of drugs over an extended period, which is beneficial for chronic conditions and long-term tissue engineering applications.

(3) Cell culture platforms: (a) Biocompatibility: Nanocellulose is non-toxic and promotes cell viability, making it an ideal material for cell culture; (b) Surface modification: The surface of nanocellulose films can be modified with peptides, proteins, or other bioactive molecules to enhance cell adhesion and proliferation. This customization enables the study of specific cell behaviors and interactions; (c) Mechanical properties: The tunable stiffness of nanocellulose films allows researchers to mimic the mechanical properties of different tissues, providing a more physiologically relevant environment for cells.

(4) Biosensing and diagnostics: (a) Electrochemical biosensors: Functionalized nanocellulose films can detect specific biomolecules, providing valuable data on tissue health and regeneration; (b) Optical biosensors: The transparency of nanocellulose films makes them suitable for optical biosensing applications, where changes in optical properties can indicate the presence of target analytes.

(5) Bioinks for 3D bioprinting: (a) 3D printed scaffolds: Nanocellulose-based bioinks can be used to print scaffolds with precise geometries and porosities, essential for tissue engineering; (b) Cell encapsulation: Cells can be encapsulated within nanocellulose matrices, allowing for the fabrication of tissue constructs with embedded living cells.

(6) Wound healing and skin regeneration: (a) Moisture retention: They maintain a moist environment, which is critical for wound healing; (b) Antibacterial properties: When combined with antimicrobial agents, nanocellulose films can prevent infections; (c) Biocompatibility: They are compatible with human skin and promote the growth of new tissue.

Nanocellulose films hold immense potential in tissue engineering applications due to their unique properties and versatility. From serving as scaffolds for tissue regeneration to acting as drug delivery systems and cell culture platforms, their applications are diverse and promising. Future research and development will likely expand their use in clinical settings, further advancing the field of tissue engineering.

Nanocellulose films have emerged as promising materials for drug delivery applications due to their unique properties, such as high surface area, biocompatibility, mechanical strength, and tunable surface chemistry. Here’s an in-depth look at the various drug-delivery applications of nanocellulose films:

(1) Controlled release systems: Nanocellulose films can be engineered to deliver drugs in a controlled manner, releasing therapeutic agents at a specific rate over an extended period. This is advantageous for maintaining optimal drug levels in the body and enhancing therapeutic efficacy; (a) Sustained release: Nanocellulose films can be designed to provide a sustained release of drugs, ensuring a consistent therapeutic effect and reducing the frequency of drug administration; (b) Triggered release: These

films can be modified to release drugs in response to specific stimuli such as pH, temperature, or the presence of certain enzymes, providing targeted therapy.

(2) Localized drug delivery: Nanocellulose films can be used for localized drug delivery, where the therapeutic agent is released directly at the site of action. This approach minimizes systemic side effects and maximizes the drug concentration at the target site; (a) Wound healing: Films impregnated with antibiotics, anti-inflammatory agents, or growth factors can be applied to wounds to promote healing and prevent infections; (b) Cancer therapy: Nanocellulose films loaded with chemotherapeutic agents can be implanted near tumor sites, delivering high concentrations of the drug directly to the cancer cells.

(3) Transdermal drug delivery: Nanocellulose films are suitable for transdermal drug delivery systems, where drugs are delivered through the skin. This method offers several benefits, including avoiding the gastrointestinal tract and first-pass metabolism, and providing a steady drug release; (a) Patches: Nanocellulose-based transdermal patches can deliver medications such as pain relievers, hormones, or nicotine in a controlled manner; (b) Enhanced permeability: The films can be engineered to enhance skin permeability, allowing for efficient drug absorption.

(4) Oral drug delivery: Nanocellulose films can be used to create oral drug delivery systems that protect drugs from the harsh environment of the gastrointestinal tract and enhance their bioavailability; (a) Encapsulation: Drugs can be encapsulated within nanocellulose matrices to protect them from degradation and release them at specific sites within the gastrointestinal tract; (b) Mucoadhesive films: These films can adhere to the mucosal lining of the gastrointestinal tract, providing a localized and prolonged drug release.

(5) Implantable drug delivery systems: Nanocellulose films can be used as part of implantable drug delivery systems, offering a controlled release of therapeutic agents over an extended period; (a) Biodegradable implants: Nanocellulose films can be incorporated into biodegradable implants that gradually release drugs as they degrade, providing long-term treatment options for chronic conditions; (b) Reservoir systems: These films can act as reservoirs that release drugs in a controlled manner, useful for delivering hormones, antibiotics, or other medications.

(6) Ocular drug delivery: Nanocellulose films can be used in ocular drug delivery systems to treat eye diseases. Their biocompatibility and ability to provide sustained release make them suitable for this sensitive application; (a) Eye drops and inserts: Nanocellulose-based formulations can be used as eye drops or ocular inserts to deliver drugs for conditions such as glaucoma, infections, or inflammation; (b) Barrier properties: The films can protect the drug from degradation while providing a controlled release to the ocular tissues.

(7) Antibacterial and antimicrobial delivery: Nanocellulose films can be loaded with antibacterial or antimicrobial agents to prevent infections, particularly in wound dressings or surgical implants; (a) Wound dressings: Nanocellulose films containing silver nanoparticles or other antimicrobial agents can be used to dress wounds, providing both protection and therapeutic benefits; (b) Catheters and medical devices: Coating medical devices with antimicrobial-loaded nanocellulose films can prevent biofilm formation and reduce the risk of infections.

Advantages of nanocellulose films in drug delivery:

(1) Biocompatibility: Nanocellulose is naturally derived and compatible with human tissues, minimizing adverse reactions.

(2) High surface area: The high surface area allows for efficient drug loading and release.

(3) Mechanical strength: Nanocellulose films are mechanically robust, providing structural integrity in various applications.

(4) Tunable properties: The surface chemistry and porosity of nanocellulose films can be easily modified to control drug release profiles.

In conclusion, nanocellulose films offer a versatile and effective platform for drug delivery applications, from controlled and localized release to transdermal and ocular delivery systems. Their unique properties make them suitable for a wide range of therapeutic applications, potentially improving treatment outcomes and patient compliance. Future research and development will likely further enhance their capabilities and expand their use in clinical settings.

12. Conclusion

The spraying of NC films offers a rapid and scalable method for fabricating free-standing films with outstanding uniformity and adjustable thickness. This technique has the potential to meet the demands of large-scale production and expand its application in various fields. Spray-coated free-standing NC films have immense potential in the biomedical field, offering opportunities for wound healing, drug delivery, tissue engineering, device coatings, and biosensing applications. Their unique properties, such as biocompatibility, high mechanical strength, porous structure, large surface area, and tailorable properties, make them promising candidates for various applications. As research in this area continues, we can expect to see even more exciting applications emerge in the future.

Disclosure statement

The author declares no conflict of interest.

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