

Nanocellulose as Sustainable Bio-Nanomaterial for Packaging and Biomedical Applications

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Abstract: Fibrous substrates made of cellulose like paper and paperboard are widely used in packaging. Furthermore, cellulose materials are biodegradable and completely safe for the environment. The water vapor barrier properties of paper and paperboard are limited due to the hydrophilic nature of cellulose fibrils. Paper is frequently combined with other materials, such as plastics, wax, and aluminum to improve its water barrier properties. Unfortunately, these materials lead to significant environmental problems and are difficult to recycle. Nanocellulose has shown the potential of being a substitute natural nanomaterial for creating environmentally friendly and sustainable barrier materials for a range of uses, including food packaging materials and substrates for the development of functional materials. Numerous cellulose sources can be used to create nanocellulose, which offers benefits like a high aspect ratio and tensile strength, making it useful for a variety of applications. The production of nanocellulose is done through a variety of processes. Potential biomedical applications of nanocellulose are tissue engineering, drug delivery, wound dressings, biosensors, implants and prosthetics, and nerve regeneration. Its application in these fields will result in the advancement of medical technology and the improvement of healthcare outcomes. Furthermore, biodegradable materials, barrier coatings, antimicrobial packaging, sustainable packaging solutions, smart packaging, and lightweight, high-strength packaging can all be made with nanocellulose. Nanocellulose-based packaging will improve the sustainability and efficiency of the packaging supply chain while providing environmentally friendly substitutes for conventional packaging materials.

Keywords: Nanocellulose; Spray coating; Vacuum filtration; Suspension consistency; Uniformity; Basis weight; Thickness

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1. Introduction

Synthetic plastics have been the foundation for the development of many functional materials in recent decades, including food packaging, flexible electronics, coatings for paper substrates, etc ^[1]. These plastics are not biodegradable, have low recycling rates, and pollute the environment when they decompose ^[2]. These materials

are currently among the major sources of soil, water, and land pollution (micro- and nano-plastic pollution)^[3]. These nano- and micro-plastics are dangerous pollutants that can infiltrate human bodies through a variety of pathways like the food chain^[4]. Biopolymers are an important source of biomaterials for the development of functional materials in the food packaging industry, and they can help alleviate these problems in the modern world^[5]. These biopolymers are typically environmentally friendly, biodegradable, and modifiable through chemical processes^[6]. The most common fibrous-based biopolymer is cellulose, which can be modified in many ways to suit a wide range of functional needs^[7]. Paper and paper boards are created using cellulose for a variety of uses^[5,8].

The most common cellulose fibrous materials are paper and paper board, which are primarily used in the packaging industry as well as bases for printed flexible electronics and packaging wrap. Paper and paperboard have large pores because they are made of cellulose microfibrils. This property allows for the easy passage of oxygen and water vapor through it. However, when exposed to high humidity, the fibers soak up water vapor, making the paper less effective as a barrier. This makes the fibrous structure wet and loose, which further enlarges the pores and weakens the material's mechanical strength. Hence, despite the biodegradability and environmental friendliness of paper and paperboard, they face limitations in terms of performance^[9].

Coating techniques have been developed to improve the mechanical and barrier qualities of paper and paperboard. These coating methods improve the mechanical characteristics, surface roughness, and barrier performance of the paper substrates by altering their surfaces. The materials used to coat paper substrates through extrusion and lamination are typically wax and synthetic plastics. Unfortunately, these materials are not recyclable and can pose a risk of contaminating the environment^[8]. Coating paper and paperboard with nanocellulose is an innovative method for enhancing their properties. Nanocellulose can fill the surface pores of the paper, effectively covering its surface and creating a barrier film. This process results in significant changes to the paper's properties compared to uncoated substrates^[5,8,10].

Nanocellulose is a type of nanomaterial composed of cellulose nanofibers and nanoparticles, created using a variety of techniques, including mechanical, chemical, and enzymatic methods, from the pulps of different cellulose sources, including wood, hemp, grass, and cotton^[11]. Nanocellulose is a carbohydrate-based renewable fibrous nanomaterial that is biodegradable and environmentally friendly^[12]. It is created through the mechanical, chemical, or enzymatic breakdown of either cellulose macrofibres or microfibers^[7,13]. Nanocellulose have an average length of 8 microns and a diameter that ranges from 5 to 100 nm^[11,14]. In addition to its use in coating paper substrates and paperboards, nanocellulose serves as a fundamental component for various applications. It acts as a building block for creating thin films used in biological applications, as well as flexible sheets employed as substrates for flexible printed electronics. Nanocellulose also finds applications in tissue engineering, composite materials, and food packaging, among other uses^[15-19].

Since nanocellulose is a renewable, recyclable, compostable, and biodegradable nanofiber, it has the potential to replace synthetic plastics^[19,20]. Due to its unique properties, free-standing nanocellulose film is developed and serves as a foundational substrate for various novel materials. These include membranes used in wastewater and water treatment, as well as membranes designed for virus removal^[17, 21-22]. Additionally, nanocellulose-based films are used as substrates in tissue engineering and flexible electronics^[16-17,23]. The methods used to coat nanocellulose onto paper substrates include traditional paper-producing processes such as roll-to-roll coating, rod coating, bar coating, and vacuum filtration. Additionally, a recently reported method involves spray coating. While solvent casting is another option for forming barrier layers on paper substrates, it is generally considered time-consuming compared to these other methods^[24]. This review demonstrates nanocellulose's diverse functionality, emphasizing its applications in biomedical and packaging sectors through

a comprehensive analysis of scientific literature.

2. Nanocellulose

The use of nanotechnology in the production of packaging materials has become increasingly popular [25]. Utilizing nanocellulose as a packaging material offers an innovative solution to replace environmentally harmful synthetic plastics [26]. Thanks to its biodegradability, biocompatibility, and strong mechanical properties, nanocellulose-based packaging materials, alongside other functional materials, are well-suited for specific applications [19]. Derived from various sources such as grass, hemp, cotton, and wood, nanocellulose is a carbohydrate-based biopolymer and nanomaterial [11]. A variety of techniques, including high-pressure homogenization, high-intensity ultrasonication, micro-grinding, micro-fluidization, electrospinning, steam explosion, and chemical processes like acid hydrolysis, enzymatic hydrolysis, and 2,2,6,6-tetramethylpiperidine 1-oxyl radical (TEMPO) oxidation, can be used to produce nanocellulose from wood and other cellulosic materials. Nanocellulose offers stability, biodegradability, and biocompatibility [27]. Research on utilizing nanocellulose to develop barrier materials for packaging applications has experienced rapid growth recently. With nanocellulose being readily available, the starting materials for packaging products made from it could be less expensive. Moreover, nanocellulose offers recyclability and reusability, paving the way for a sustainable approach to packaging [28,29].

Nanocellulose are characterized by their nanometer-scale fiber diameter and width [11]. The delamination process involves breaking down cellulose fibers into microfibrils or nanofibers, typically sourced from wood or cotton pulps [30]. Micro-fibrillated cellulose, for instance, has an aspect ratio of 142, with an average fiber length of 8 μm . Nanocellulose forms a fibrous matrix composed of entangled cellulose nanofibers and microfibrils of various sizes and distributions [12]. The hierarchy of cellulose fibers from wood to nanofibers is shown in **Figure 1**. **Figure 1** illustrates the process by which nanofibers are separated from cellulose macrofibres using a variety of chemical and mechanical techniques, including ball milling, homogenization, and acid hydrolysis.

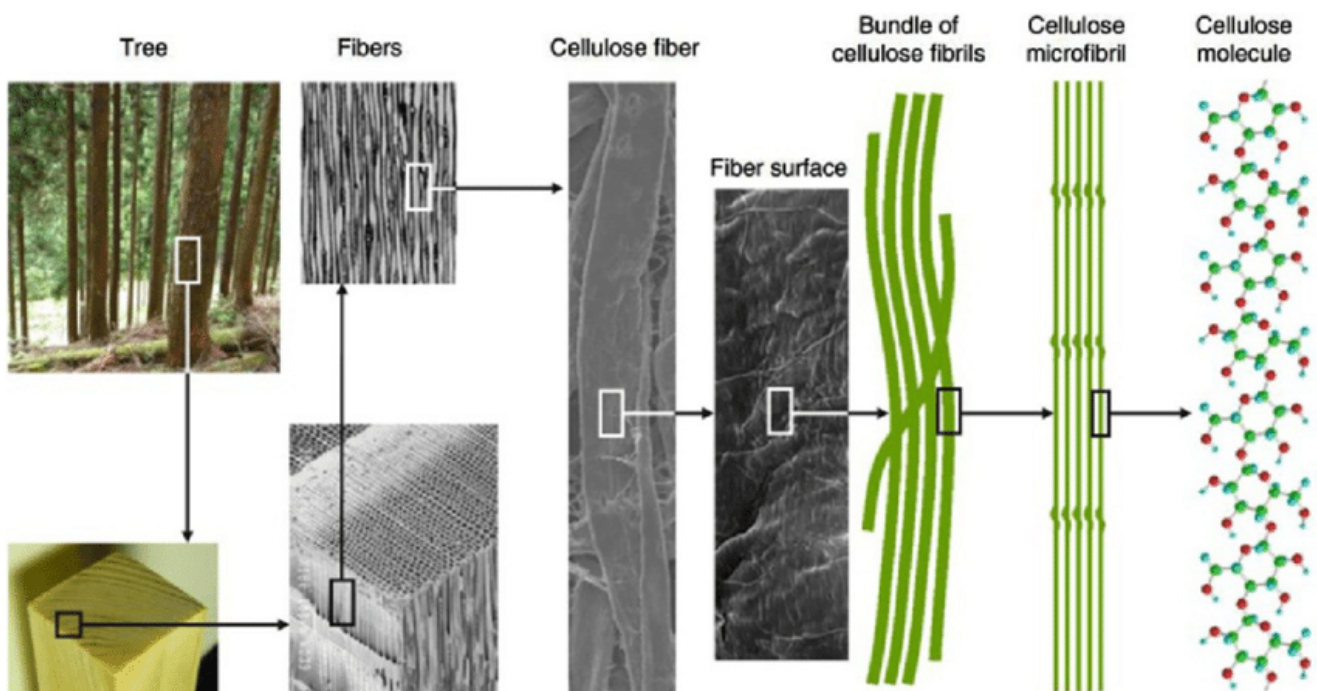


Figure 1. Hierarchy of cellulose fibers from wood to nanofibers/nanocellulose [31]

3. Characteristics of nanocellulose

There are two types of arrangements of these long, flexible cellulose fibers: amorphous and crystalline. The distribution and arrangement of these fibers play a crucial role in determining various functional properties, especially the barrier performance of nanocellulose coatings and films. **Figures 2 and 3** illustrate the surface morphology of nanocellulose after two cycles of high-pressure homogenization. Following high-pressure homogenization, the diameter of nanocellulose is estimated to be around 20 nm. The diameter and length of nanocellulose contribute to their high tensile strength (10 GPa) and elastic modulus (150 GPa). These characteristics, along with the diameter and aspect ratio of the nanofibers, are influenced by factors such as the source of cellulose, processing techniques, and the type of particles used in the feedstock. These parameters dictate the applicability and functionality of nanocellulose in various applications [7].

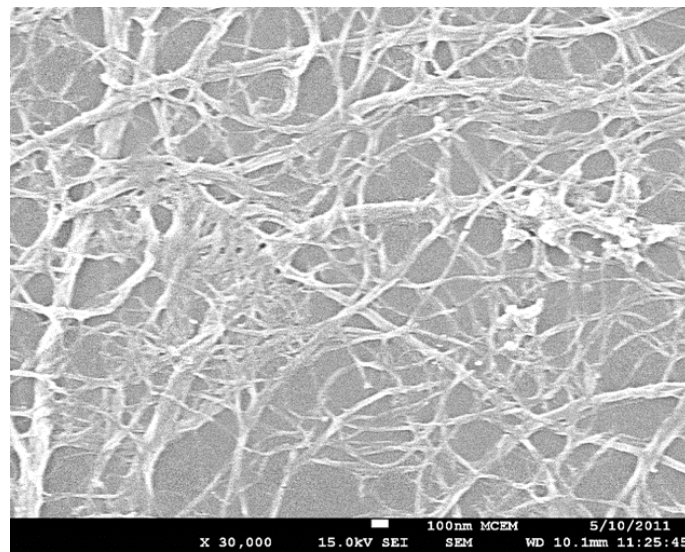


Figure 2. Surface morphology of nanocellulose after two cycles of high-pressure homogenization I

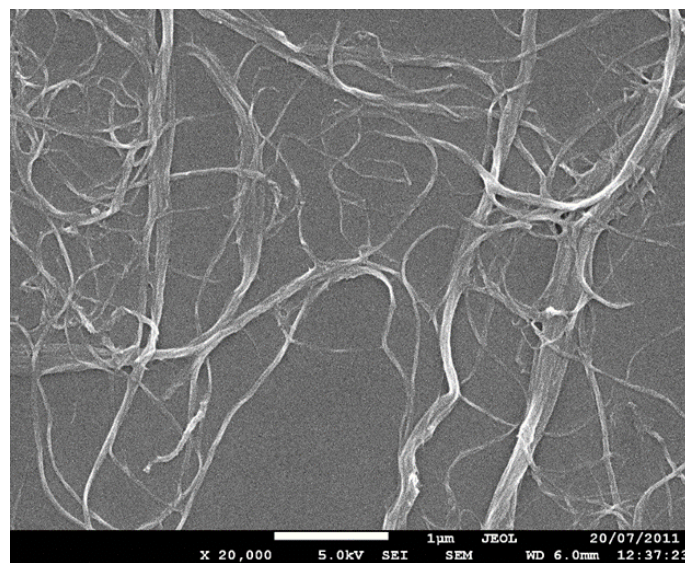


Figure 3. Surface morphology of nanocellulose after two cycles of high-pressure homogenization II

Following high-pressure homogenization, the nanocellulose are entangled to form a fibrous mesh, as confirmed by the scanning electron microscope (SEM) micrographs. Furthermore, the size distribution of the nanofibers varies, and some of them aggregate due to the formation of hydrogen bonds between adjacent hydroxyl groups. The size distribution of nanofibers in nanocellulose is depicted in **Figure 4**, which confirms that the majority of the nanofibers' sizes fall below 100 nm and that their average diameter is estimated to be 73 nm. Due to aggregation, some nanocellulose exhibit sizes greater than 100 nm ^[24,30].

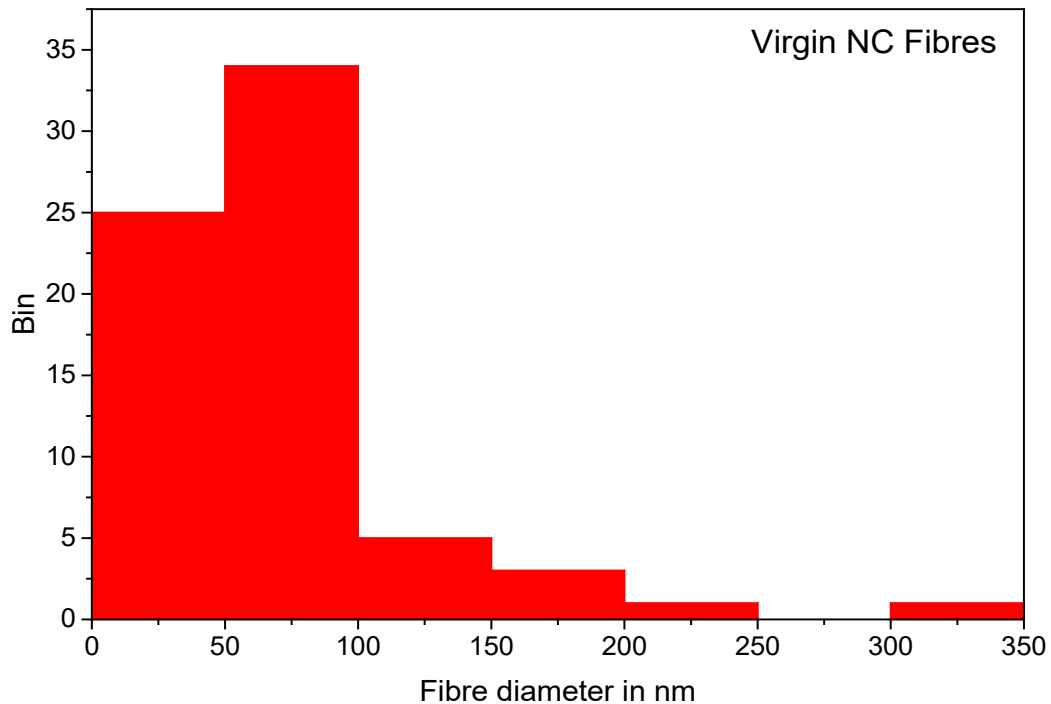


Figure 4. Size distribution of nanocellulose

Nanocellulose typically have a diameter of 5 nm to 100 nm and a length of several microns per fibril. The structure of the fiber chain is usually both crystalline and amorphous ^[32]. The functionality of the material is typically attributed to the crystalline region. For example, in films, the presence of crystalline areas creates a convoluted pathway that enhances the barrier performance of nanocellulose films. This expansion of the diffusion pathway improves the barrier properties against gaseous molecules like oxygen and water vapor. Additionally, fiber diameter and length indirectly influence the rheological and interfacial properties of nanocellulose suspension. The distribution of nanocellulose diameter and aspect ratio of nanocellulose depends on various factors, including the source, pre-treatment processing, and fibrillation processes ^[24,30].

The SEM, transmission electron microscopy (TEM), and atomic force microscopy (AFM) micrographs were analyzed to determine the diameter and aspect ratio of the nanocellulose. The gel point of nanocellulose fibers can be determined using the sedimentation method. This point marks the transition from a low-concentration nanocellulose suspension to a dense, thick, concentrated suspension, where hydrogen bonds among the nanofibers cause them to form a compact network. This network, known as the threshold of connectivity, signifies the onset of gelation in the suspension. The suspension becomes weaker after this concentration because of insufficient contact between the fibers. The aspect ratio of nanocellulose fibers influences this property, which in turn affects the vacuum filtration drainage time required for creating nanocellulose films ^[27].

Lightweight, optical transparency, modifiability, dimensional stability, and good barrier properties are some of nanocellulose's noteworthy characteristics. When creating scaffolds for tissue engineering and drug delivery/targeting vehicles, nanocellulose works incredibly well with other materials such as proteins, natural polymers, and living cells. The hydroxyl groups of the nanocellulose surface allow for functionalization and chemical modification, such as the addition of amine and hydroxy terminals, which can increase the binding targets' affinity for the polymers. A critical requirement for coating nanocellulose on paper substrates and other surfaces is the viscosity of the nanocellulose suspension. Nanocellulose suspension typically displays non-Newtonian fluid behavior, resembling the shear-thinning behavior observed in pseudoplastic fluids ^[24].

4. Production of nanocellulose

The feedstocks used to produce nanocellulose include wood, seed fibers (cotton, coir, etc.), bast fibers (flax, hemp, jute, ramie, etc.), and grasses (bagasse, bamboo, etc.). Carrots and bagasse have been used as sources for nanocellulose production in recent years. In order to produce nanocellulose, recycled cellulose fibers, such as waste paper, are homogenized. Numerous studies are being conducted to develop inexpensive feedstock. On the other hand, microorganisms such as bacteria, fungi, algae, and tunicates are not given as much thought in terms of producing nanocellulose ^[33].

One of the most crucial feedstocks for nanocellulose production is the wood pulp. The type of wood used can be soft or hardwood. The structural variation in the wood's cellulose fiber hierarchy is what distinguishes hardwood from softwood. The primary component of wood is cellulose; other constituents include hemicellulose, lignin, and some other inorganic materials. Hardwood is typically more rigid than softwood. Softwood is preferred for the production of nanocellulose because it is easier to homogenize or defibrillate. Similarly, less energy is needed to produce nanocellulose from non-woody cellulose feedstock like grass, carrots, and marine sources. Lignin contamination is a common issue when using wood as a feedstock for nanocellulose. Thus, the nanocellulose that is produced from wood needs to be further purified to remove all traces of lignin. Food and agricultural waste have recently been utilized as a feedstock for the production of nanocellulose. When free of other cellulose materials like hemicelluloses and lignin, nanocellulose fibrillation requires less energy. The difficult stage in the fibrillation process is refining, which is the process that turns cellulose microfibrils into cellulose nanofibers or nanocellulose. Refining can be performed in three ways: enzymatic, chemical, and mechanical refining. The microfibers must be purified (delignified) before being refined. This lowers the energy needed for fibrillation processes and yields high-quality nanocellulose.

One of the most crucial steps in the mechanical fibrillation of cellulose microfibers into cellulose nanofibers is disintegration. Grinding is the most effective technique for producing nanocellulose. High-pressure homogenization is a novel technique for fibrillating microfibers into nanofibers. However, this method has high energy consumption — 25,000 kWh per ton of microfibers. The mechanical method works well for commercialization and large-scale production. The process' energy consumption is primarily determined by the pressure, nanocellulose suspension concentration, and number of passes in the high-pressure homogenization method. **Table 1** lists the different techniques for breaking down fibers mechanically. It provides an overview of the reduction of cellulosic fibers, the benefits and drawbacks, isolation techniques, and the mechanism of each method ^[7].

Table 1. Mechanical refining for the production of nanocellulose ^[7]

Methods for isolating cellulosic fibers	Mechanism of size reduction	Advantages	Disadvantages
High-pressure homogenization	Reduces fiber size by high-impact shearing forces	(1) It is a quick, effective, and continuous process. (2) It has great reproducibility and can control the degree of defibrillation	(1) Clogging (2) Requires pre-treatment of fibers (3) High passing time (4) High energy consumption (5) Increases temperature of suspension
Micro-fluidization	High-impact collision leading to the splitting of macrofibres into nanofibers/nanocellulose	(1) Less clogging (2) Uniformity in size (3) Fewer cycles	Not suitable for large-scale production
Micro-grinding	The microfibrils are pressed between the stator and the rotor disk of the grinder. The high frictional forces tear the fibers into fibrils	(1) Lower energy consumption and fewer cycles (2) No pre-treatment is required	(1) High maintenance cost (2) Difficulty in replacing internal parts like the disc (3) Reduces the crystallinity of the nanocellulose
High-intensity ultrasonication	Microfibers are disintegrated into nanofibers using sound energy	(1) High power output (2) Efficient defibrillation	(1) Generates heat and noise (2) Requires pre-treatment (3) Only applicable in laboratories
Refining	Microfibers are disintegrated through high shear forces		
Cryo-crushing	The refined macrofibers are treated with liquid nitrogen and ground.	High disintegration performance	Formation of ice
Steam explosion	The suspension is heated at high pressure and then vented into a vessel with low pressure.	-	Requires chemical pre-treatment

As mentioned earlier, a significant hurdle in scaling up and commercializing nanocellulose production is the energy-intensive mechanical fibrillation process, which converts microfibrils into nanofibers. The demand for cellulosic nanomaterials has surged in various industries and research fields, driving the need for large-scale production. Therefore, scaling up nanocellulose production requires the development of an energy-efficient process ^[30]. When compared to mechanical disintegration processes, chemical or enzymatic processes have been shown to require less energy and have higher efficiency ^[7]. **Table 2** lists several cellulosic nanofiber chemical processing methods along with their benefits and drawbacks.

Table 2. Chemical processing for Nanocellulose ^[30]

Chemical method	Reaction mechanism	Advantages	Disadvantages
TEMPO-oxidation	Oxidation of the C6 hydroxyl groups into carboxyl groups and partially into aldehydes	Shorter reaction time	TEMPO is toxic and expensive
Periodate chlorite oxidation	Oxidation of the vicinal hydroxyl groups in the C2 and C3 positions	Increases carboxyl groups	(1) Weakens the structure of cellulose (2) Long reaction time
Alkaline extraction	The fibrils are pretreated before chemical processing	(1) Degrades lignin (2) Enhances fibrillation	(1) Degradation of cellulose (2) High alkali concentration
Carboxymethylation	Carboxymethylated groups are introduced to the fibers, which enhances the anionic groups and reduces the friction between the fibers, which in turn facilitates disintegration	(1) Improves fibrillation (2) Low energy consumption	Produces thinner NFC
Acid hydrolysis	Works by breaking the glycoside bonds	High crystallinity of nanocellulose	(1) Uses strong mineral acids (2) Requires temperature control

Enzymatic procedures are used to turn cellulose biomass into nanocellulose. This technique involves breaking the glycosidic bonds in the cellulose molecules through enzymatic hydrolysis. The two most common enzymes used in producing nanocellulose are cellobiohydrolase and endoglucanase; the former targets the crystalline portion of cellulose, while the latter targets the amorphous portion of cellulose. The length of the hydrolysis process, the concentration of enzymes in the reaction mixture, and the type of enzymes used all affect the amount of nanocellulose produced. Chemical and enzymatic processes are typically carried out before mechanical processes to prevent clogging and reduce energy consumption. This leads to efficient fibrillation and the production of high-quality fibers^[27,34-35].

5. Biomedical application of nanocellulose

Nanocellulose, which is derived from cellulose fibers, has a wide range of biomedical applications due to its unique properties such as high surface area and mechanical strength, and biocompatibility. Below are some of the biomedical applications of nanocellulose.

(1) Tissue engineering

Nanocellulose has been researched for its use in creating scaffolds for tissue engineering. Its biocompatible nature and ability to mimic the structure of natural extracellular matrix make it a promising material for promoting cell growth and tissue regeneration.

(2) Wound dressings

Nanocellulose-based wound dressings have been developed, with the ability to absorb exudate, maintain a moist environment, and promote wound healing. The large surface area of nanocellulose also enables the loading of bioactive compounds for enhanced healing properties.

(3) Drug delivery

Nanocellulose can be used as a carrier for drug delivery due to its ability to encapsulate and release drugs in a controlled manner. Its biodegradability and low toxicity also make it a suitable material for targeted drug delivery.

(4) Biosensors

Nanocellulose-based biosensors have been developed for applications such as glucose monitoring and the detection of biomolecules. The high surface area and unique mechanical properties of nanocellulose make it suitable for creating sensitive and selective biosensors.

(5) Implants and prosthetics

Nanocellulose has been explored for the development of implants and prosthetics due to its mechanical strength and biocompatibility. It could potentially be used to create bone scaffolds, joint replacements, and other biomedical implants.

(6) Nerve regeneration

Nanocellulose has shown potential in promoting and supporting neural cell growth. Overall, nanocellulose plays a significant role in improving healthcare outcomes and advancing medical technology. Continued research and development in this area are anticipated to result in the commercialization of nanocellulose-based biomedical products in the near future.

6. Application of nanocellulose in packaging

Fibrous substrates made of cellulose like paper and paperboard are widely used in packaging. Furthermore, cellulose materials are biodegradable and completely safe for the environment. The water vapor barrier

properties of paper and paperboard are limited due to the hydrophilic nature of cellulose fibrils. Paper is frequently combined with other materials, such as plastics, wax, and aluminum to improve its water barrier properties. Unfortunately, these materials lead to significant environmental problems and are difficult to recycle. Nanocellulose has shown the potential of being a substitute natural nanomaterial for creating environmentally friendly and sustainable barrier materials for a range of uses, including food packaging materials and substrates for the development of functional materials. Numerous cellulose sources can be used to create nanocellulose, which offers benefits like a high aspect ratio and tensile strength, making it useful for a variety of applications. The production of nanocellulose is done through a variety of processes. Potential biomedical applications of nanocellulose are tissue engineering, drug delivery, wound dressings, biosensors, implants and prosthetics, and nerve regeneration.

(1) Biodegradable packaging materials

Nanocellulose can be used to create biodegradable packaging materials such as films, coatings, and trays. It is an eco-friendly alternative to traditional plastic packaging.

(2) Barrier coatings

Nanocellulose can be used as a barrier coating for packaging to improve the moisture resistance and gas barrier properties of the packaging material. This can help extend the shelf life of food products and reduce the need for additional layers of packaging.

(3) Antimicrobial packaging

Nanocellulose can be functionalized with antimicrobial agents to create packaging materials that help extend the shelf life of perishable products by inhibiting the growth of bacteria and fungi.

(4) Sustainable packaging solutions

Nanocellulose-based materials can be used to create sustainable and environmentally friendly packaging. It can be produced from renewable resources and offers improved recyclability and compostability compared to traditional packaging materials.

(5) Smart packaging

Nanocellulose can be used to create smart packaging that incorporates sensors or indicators to monitor the quality and freshness of the packaged products. This can help reduce food waste and ensure product safety.

(6) Lightweight and high-strength packaging

Nanocellulose-based materials offer high strength-to-weight ratios, making them suitable for making lightweight yet durable packaging. This can help reduce transportation costs and improve the overall efficiency of the packaging supply chain.

7. Conclusion

Synthetic plastics are a major source of pollution and environmental harm, but nanocellulose, a renewable and biodegradable nanomaterial, is a promising replacement for synthetic plastics due to its biodegradability, biocompatibility, and high mechanical strength^[35]. Nanocellulose can be produced from various cellulose sources and has unique characteristics such as high tensile strength and aspect ratio, making it suitable for a wide range of applications. Nanocellulose can be manufactured in many ways. It also has a wide range of potential biomedical applications, including tissue engineering, wound dressings, drug delivery, biosensors, implants and prosthetics, and nerve regeneration. It plays an important role in improving healthcare outcomes and advancing medical technology. Additionally, nanocellulose can be used to create biodegradable, antimicrobial packaging, sustainable, smart, lightweight, and high-strength packaging. This material is an

environmentally friendly alternative to traditional packaging materials and can improve the efficiency and sustainability of the packaging supply chain.

Disclosure statement

The authors declare no conflict of interest.

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