Book Chapter

Nanocellulose and Its Potential in the Paper and Packaging Industry

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1. Introduction

Nanocellulose, derived from various plant sources, has gained significant attention in recent years due to its unique properties and versatile applications across multiple fields, including materials science, biomedical engineering, environmental applications, electronics, energy, etc. The production of nanocellulose is a rapidly evolving field that leverages innovative techniques and advanced practices. It involves a combination of mechanical, chemical, and enzymatic methods, each contributing

to the remarkable physical, mechanical, biological as well as chemical properties of the final product such as large specific surface area, biocompatibility, rapid biodegradability, low toxicity, excellent strength properties and, last but not least, extensive chemical modification possibilities. Using agricultural residues as feedstocks presents a sustainable approach to nanocellulose production, aligning with the growing demand for eco-friendly materials. The ongoing research and development efforts are essential for optimizing production processes and expanding the full potential of nanocellulose in a wide range of industries.

This book chapter aims to first introduce nanocellulose in general, i.e. sources, production, types, properties, and applications, and then focus on its use in the paper and packaging industry. The application of nanocellulose in papermaking represents a significant advancement in material science, offering enhanced mechanical properties, improved barrier functions, and sustainable production practices. The contribution emphasizes the potential of nanocellulose to revolutionize traditional papermaking processes and products.

2. Cellulose as a Source of Nanocellulose

Cellulose is the most common organic polymer and a major component of the cell walls of plants, representing about 1.5 trillion tons of the total annual biomass production [1]. It is a complex carbohydrate made up of long chains of D-glucopyranose units linked together by β(1→4) glycosidic bonds (Figure 1). This linear stiff-chain homopolymer is characterized by its hydrophilicity, chirality, biodegradability, broad chemical modifying capacity, and its formation of versatile semi-crystalline fiber morphologies. Cellulose fibers give plants structural strength and rigidity [2,3].

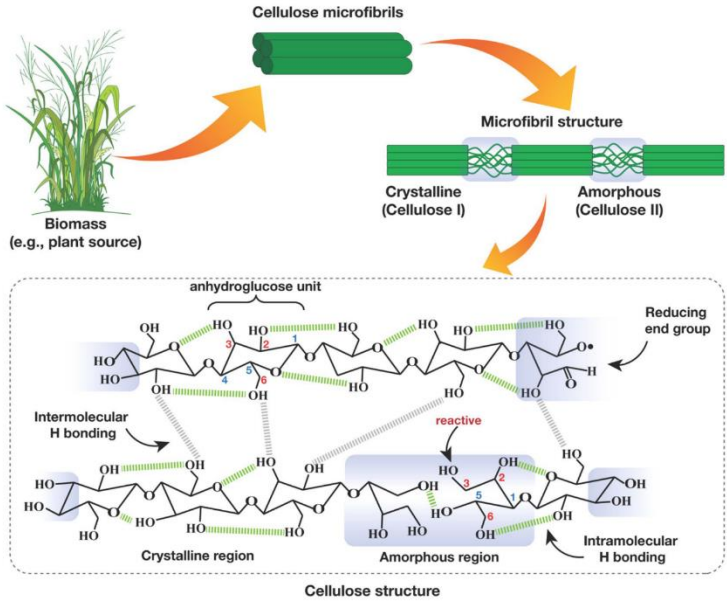

Figure 1: Molecular structure of cellulose obtained from biomass. Cellulose microstructure contains less reactive compact crystalline and highly reactive amorphous regions [4]

The average degree of polymerization (DP) varies considerably among cellulose samples. In the native form, DP is 6,000–14,000, while in cellulose isolated from wood that has undergone some technological process, it decreases, and in most cases, it ranges from 500 to 2,500 [2].

Cellulose does not dissolve in water and common organic solvents but swells only to a limited extent. This is mainly due to strong intermolecular hydrogen bonds, which significantly contribute to forming a specific supramolecular cellulose structure. In concentrated acids, cellulose dissolves while reducing the average DP. These reactions are often used in various analyses of cellulose [5]

The stabilization of cellulose macromolecular chains in an ordered system, i.e. its supramolecular structure, is ensured by functional groups capable of interacting. These cellulose hydroxyl (–OH) groups can form two types of hydrogen bonds, depending on their

position in the glucose molecule. In the first case, hydrogen bonds are formed between the –OH groups of adjacent glucose units in one cellulose macromolecule (intramolecular hydrogen bonds). These bonds cause less flexibility in the cellulose chain. The second type of hydrogen bonds are bonds between the –OH groups of adjacent cellulose macromolecules (intermolecular hydrogen bonds), which participate in the formation of the supramolecular structure of cellulose [6,7].

The simultaneous application of hydrogen bonds and van der Waals interactions at a certain approximation of linear cellulose chains determines the formation of the crystalline structure of cellulose. Native cellulose can contain up to 70% of ordered crystalline regions, the remaining part is composed of amorphous parts. The mutual ratio depends on the origin and isolation of cellulose. It is always a semi-crystalline polymer. The degree of crystallinity, the type of crystalline modification, and the length of cellulose chains affect the physicochemical properties of cellulose and its reactivity (e.g. the degree of wood swelling, chemical resistance, bending, brittleness, etc.) Crystalline regions are much more stable and most chemical reactions take place mainly in amorphous parts, which are more accessible to reagents. Cellulose in its native state is referred to, according to its crystal lattice, as cellulose I. It is a mixture of cellulose Iα and Iβ in a ratio depending on the plant species. Iβ is typical of higher plants, including wood, and its lattice is monoclinic. Iα has a triclinic crystal lattice, as shown in Figure 2 [2].

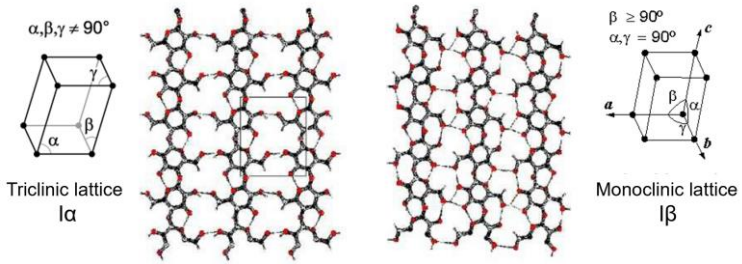

Figure 2: Triclinic (Iα) and monoclinic (Iβ) crystal lattice of cellulose I [2]

Currently, the largest source of industrialized cellulose, or more precisely pulp, is wood, but it occurs naturally in all plants [8]. In addition to wood and plants, the source of cellulose can also be animals, algae, or bacteria [9]. The source of so-called "green" nanocellulose is industrial waste, e.g. from beer production or solid municipal waste [10].

The potential of cellulose is tremendous. It is considered an almost inexhaustible raw material source for the increasing demand for environmentally friendly and biocompatible products. Renewable materials are very important for carbon capture and sequestration, which prevents large amounts of carbon dioxide from being released into the atmosphere to reduce global warming [7]. Cellulose plays a crucial role in plant biology and has numerous practical applications, e.g. paper, textile, various biodegradable plastics, pharmacy, food industry, etc.

3. Nanocellulose Production – Techniques and Raw Materials

As already mentioned, nanocellulose can be derived from various sources, including wood, agricultural residues, and other lignocellulosic materials. The production methods can be broadly categorized into mechanical, chemical, and enzymatic processes (Figure 3), each with its advantages and limitations.

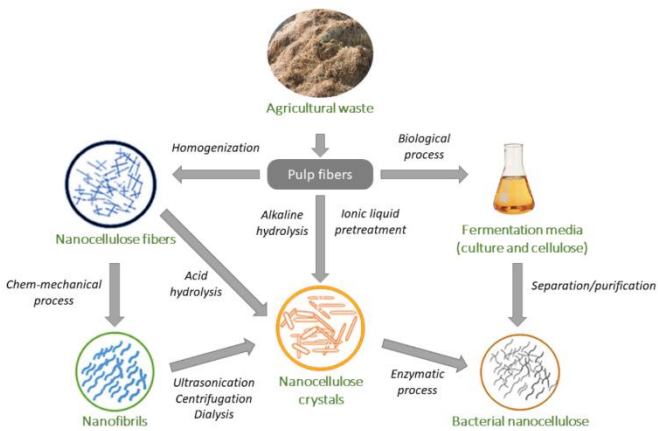

Figure 3: Schematic representation of nanocellulose production from agricultural wastes [11].

Mechanical methods, such as high-pressure homogenization and grinding, are commonly employed to produce nanocellulose fibers. These methods involve applying mechanical forces to break down cellulose fibers into nanoscale dimensions. For instance, Mittal et al. [12] demonstrated that the double flow-focusing method could produce cellulose nanofibrils (CNFs), also called nanofibrillated cellulose (NFC), with remarkable mechanical properties, making them suitable for load-bearing applications. However, these mechanical processes can be mostly energy-intensive and may lead to fiber damage if not carefully controlled [13]. Another innovative approach to nanocellulose production is the use of hydrothermal microwave-assisted techniques, as described by Melo et al. [14]. This method offers environmental and economic advantages by utilizing biomass, water, and microwave energy as inputs, leading to high-value outputs such as nanocellulose and sugar liquor. The study emphasizes the sustainability aspect of this production method, which aligns with the growing demand for eco-friendly materials in various industries.

Chemical methods, particularly acid hydrolysis, are widely used for nanocellulose production. This process selectively removes amorphous regions of cellulose, resulting in cellulose nanocrystals (CNCs) with high crystallinity. Sumarago [15] highlighted that acid hydrolysis is one of the most effective methods for producing nanocellulose, yielding fibers with enhanced mechanical properties. Trilokesh and Uppuluri [16] demonstrated that acid hydrolysis of cellulose fibers can yield spherical cellulose nanocrystals (SCNCs), which possess unique structural characteristics. Additionally, the use of TEMPO-mediated oxidation has been shown to improve the disintegration of cellulose fibers, facilitating the production of nanofibrils [17]. Kocatürk [18] utilized the TEMPO oxidation method to produce nanocellulose from bleached softwood pulp, resulting in thread-like particles with a diameter of approximately 20 nm. This method not only enhances the mechanical properties of the nanocellulose but also allows for functionalization, which can further expand its application potential.

Enzymatic methods have emerged as a promising alternative for producing nanocellulose, offering a more environmentally friendly approach compared to traditional chemical methods. Hu et al. [19] explored the synergistic effects of various enzymes, including endoglucanase and lytic polysaccharide monooxygenase, in promoting cellulose fibrillation. This enzymatic treatment can enhance the thermostability of nanocellulose, making it suitable for various applications. Furthermore, the combination of enzymatic hydrolysis with mild mechanical treatment has been shown to facilitate the production of high-solid nanocellulose, reducing energy consumption during processing [13,20].

The choice of raw materials for nanocellulose production is crucial, as it influences the properties of the final product. Agricultural residues, such as sugarcane bagasse and oil palm empty fruit bunch, have been identified as promising sources for nanocellulose extraction.

The study by Gond et al. [21] presented the effectiveness of chemical treatments in extracting nanocellulose from sugarcane bagasse, showcasing the potential of agricultural waste as a feedstock for nanocellulose production. Similarly, Charoensopa [22] highlighted the successful extraction of nanocellulose from sugarcane bagasse, emphasizing its potential for various applications, including antimicrobial materials. Zailuddin et al. [23] reported that the isolation of nanocellulose from an oil palm empty fruit bunch resulted in fibers with an average size of less than 50 nm, demonstrating the feasibility of utilizing agricultural waste for nanocellulose production. Fahma et al. [24] investigated the extraction of nanocellulose from palm oil by-products, demonstrating its biocompatibility and potential for use in textile applications. Mariño et al. [25] focused on extracting nanocellulose from citrus waste and pointed out its versatility and potential for high-performance composite applications. Mahardika et al. [26] reported that high-shear homogenization and ultrasonication of pineapple leaf fibers resulted in nanocellulose with high purity and specific dimensions, highlighting the efficiency of these methods in producing nanocellulose with desirable properties. These studies underscore

the importance of utilizing renewable resources and waste materials in the production of nanocellulose, contributing to a more sustainable circular economy.

4. Nanocellulose Types and Characterization

Nanocellulose is defined as a fibrous cellulose-based material that does not exceed 100 nm in size in at least one dimension. In general, nanocellulose can be divided into three groups according to the production method (Figure 4): cellulose nanocrystals (CNCs), rod-shaped highly crystalline cellulose nanocrystals generally obtained by hydrolysis with concentrated mineral acids (mainly HCl or H_2SO_4); cellulose nanofibrils (CNFs), usually obtained through enzymatic and/or mechanical disintegration processes; and bacterial nanocellulose (BNC), obtained almost exclusively by a family of bacteria known as *Gluconoacetobacter xylinius* [8,11,27,28].

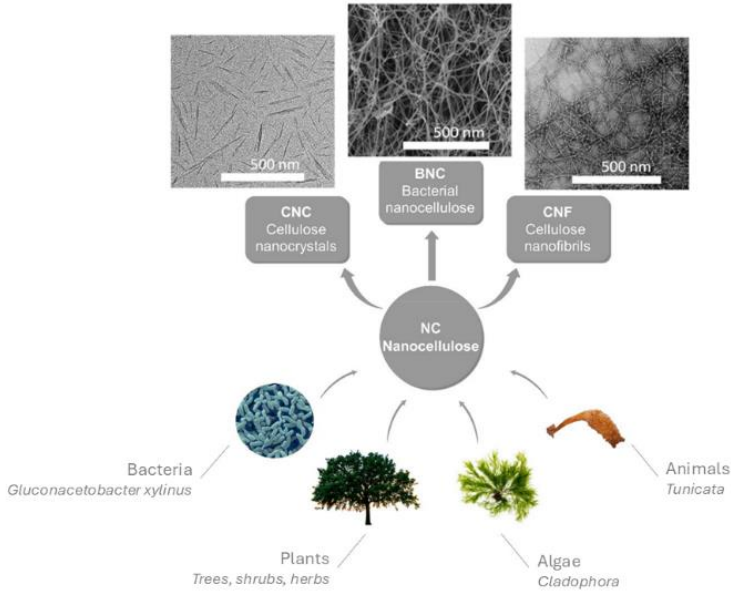

Figure 4: Different sources and types of nanocellulose and their scanning electron microscopy (SEM) images [7,29]

According to the structure, nanocellulose is divided into nanofibrils, nanofibers, nanocrystals, nanorods, nanospheres, and nanoplatelets [9]. The types of nanocellulose, their sizes, and sources are listed in Table 1.

Table 1: Nanocellulose types [10,30,31]

Nanocellulose structures	**Example**	**Dimensions**
Nanofibrils	In bacterial cellulose	Diameter from 70 to 140 nm, length in μm
	In wood-derived cellulose	Diameter 3–5 nm, length several μm, form 20–50 nm thick aggregates
Nanofibers	Created by electrospinning	Cellulose acetate: average diameter about 400 nm
		Bacterial cellulose (33 wt.%) with chitosan: diameters from 80 to 170 nm
	Isolated from pineapple	Width 6.4 ± 4.6 nm, length in μm
Nanowhiskers	Pine kraft pulp	Diameter 5–15 nm, length 100–250 nm
	Kenaf bast	Diameter 10–15 nm, length hundreds nm
	Bacterial cellulose	Diameter 10–100 nm, length 100–1000 nm
Nanocrystals	Cotton-derived	Mean width 7.3 nm, mean length 135 nm
Nanorods	Grass-derived	Width 15 ± 3 nm, length 120 ± 15 nm
Nanoballs	Wood-derived	Diameter 80–85 nm
Nanoplates	Agave-derived	Thickness 80 nm, other dimensions in μm

The term “nanofibers” is used for structures that are longer and thicker than nanofibrils. These are submicron-scale fibers and are primarily structures formed by electrostatic spinning of cellulose without additives or in composites with natural or synthetic polymers. Cellulose nanocrystals, nanofibers, and nanorods are produced by acid hydrolysis, which destroys the amorphous regions. As a result, these structures have a higher crystallinity index than cellulose nanofibrils, which are more often produced by enzymatic hydrolysis by xylanases and cellulases or by

mechanical decomposition of cellulose by grinding, homogenization, and shearing. Due to the cost of the process, chemical or enzymatic pretreatment is often performed. Nanofibrils can be further assembled into 2D and 3D microstructures and macrostructures [10,32]. CNFs obtained by mechanical processing have an intricate network structure formed by flexible nanofibers with a width of 20–100 nm and a length greater than 10,000 nm. Bacterial or microbial nanocellulose is a very promising material for biomedical use. It is composed of very fine nanofibers with a diameter of 20–100 nm, which form three-dimensional networks [9]. It is produced by gram-negative bacteria of various genera, the most important of which is the already-mentioned *Gluconoacetobacter xylinius*. Nanocellulose production can be increased by the presence of yeast. Bacterial cellulose is chemically identical to plant cellulose, but it does not contain by-products such as lignin, pectin, and hemicelluloses [33]. Its synthesis is very expensive.

5. Nanocellulose Properties

Nanocellulose is a very advantageous material in terms of physical, chemical, and biological properties. A large specific surface area, high strength, barrier properties, antimicrobial effects, and low immunogenicity generally characterize cellulose-based materials. They are chemically inert, biocompatible, and mostly non-toxic. They also have customizable physical, chemical, morphological, thermal, electrical, and optical properties. These materials are available and, above all, renewable [10,33,34].

One of the most notable properties of nanocellulose is its exceptional mechanical strength. Nanocellulose fibers, including cellulose nanofibrils (CNFs) and cellulose nanocrystals (CNCs), demonstrate high tensile strength and stiffness, often exceeding that of conventional materials. In addition, compared to metals, they are characterized by a much lower weight - the density of nanocellulose is about 1.6 g/cm^3 [27]. For instance, Gan et al. [35] and Li et al. [34] reported that nanocellulose exhibits a modulus of elasticity of up to 150 GPa, which is significant for reinforcing materials in composites. Hashemzehi et al. [36] published that nanocellulose has an elastic modulus of 110–220 GPa and a

tensile strength of 7.5–7.7 GPa. Sharma et al. [8] reported a tensile strength of 10 GPa comparable to cast iron. This high mechanical performance is attributed to the crystalline structure of nanocellulose and the dense hydrogen bonding between cellulose chains, which allows for efficient load transfer at the nanoscale [37-39].

In addition to mechanical properties, nanocellulose has a high specific surface area of up to a hundred m^2/g, enhancing its reactivity and interaction with different atoms, molecules, microbial cells, and other materials [9]. Hashemzehi et al. [36] mention a high concentration of hydroxyl groups on nanocellulose chains. This property is crucial for applications in composite materials, where nanocellulose can improve the interfacial bonding between the matrix and the reinforcement [39]. The high surface area also contributes to its effectiveness as a filler in various applications, including paper and packaging materials, where it can enhance mechanical properties such as tensile strength and elongation [40,41].

Thermal stability is another important property of nanocellulose. Studies have shown that nanocellulose can maintain its structural integrity at elevated temperatures, making it suitable for applications that require thermal resistance. For example, Zhao et al. [42] highlighted that nanocellulose-based composites exhibit improved thermal properties compared to conventional materials, which is beneficial for applications in the automotive and aerospace industries. Li et al. [34] indicate a degradation temperature of nanocellulose of around 350 °C. Trache et al. [9] and Kumar et al. [33] reported for nanocellulose a coefficient of thermal expansion of 0.01 ppm/K.

Furthermore, nanocellulose is characterized by its biodegradability and non-toxic nature, making it an environmentally friendly alternative to synthetic materials. This property is particularly advantageous in the biomedical field, where biocompatibility is essential. Wiegand et al. [43] demonstrated that bacterial nanocellulose (BNC) can be functionalized for antimicrobial applications, showcasing its potential in wound dressings and other medical devices. The biodegradability of nanocellulose aligns with the growing

demand for sustainable materials in various sectors, including packaging and construction [39].

Nanocellulose also exhibits unique optical properties, such as high transparency and low haze, which are valuable in applications like coatings and films. Abdel-Karim et al. [86] noted that the films produced from nanofibrillated cellulose demonstrate excellent optical clarity, making them suitable for use in electronic displays and packaging materials.

Last but not least, the versatility of nanocellulose allows for various chemical modifications, enhancing its functionality for specific applications. For instance, Lee and Bismarck [44] discussed green chemical modifications that can tailor the properties of nanocellulose for use in composites, improving compatibility with different matrices and expanding its application range.

Kumar et al. [33] further mention the gas impermeability, electrical conductivity, and UV-blocking properties of nanocellulose.

These properties, which are schematically summarized in Figure 5, may vary in part as they are influenced by factors such as their source, extraction method, and processing conditions.

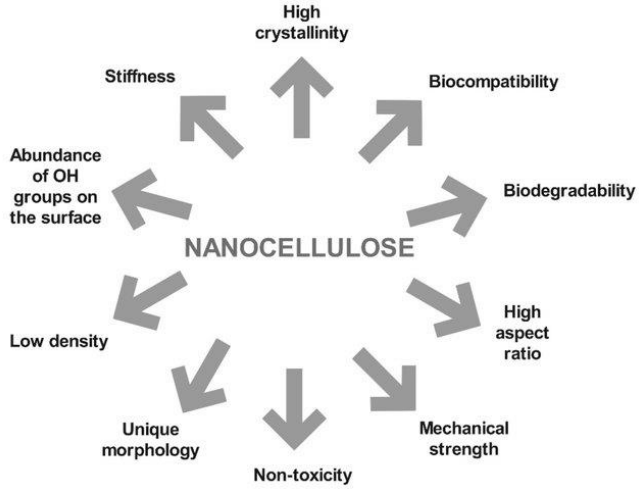

Figure 5 Unique properties of nanocellulose [29]

6. Nanocellulose Application

Due to its many advantageous properties, nanocellulose is a promising material used in many sectors such as industry, technology, and biomedicine (Figure 6). It is mainly used in air and aqueous solution purification [45-47,91], filtration [48-50], packaging [51,52], conservation of historical objects [53], energy generation and storage [54-56], as a component of flame retardants [57-59] and thermal insulators [91], in acoustics [60], sensors [61-64,91,92] and in controlled drug delivery [65-68]. In addition to these applications, Trache et al. [9] mention use in the automotive and aerospace industries, adhesives, hydrogels, nanocarriers, cementitious materials, and defense.

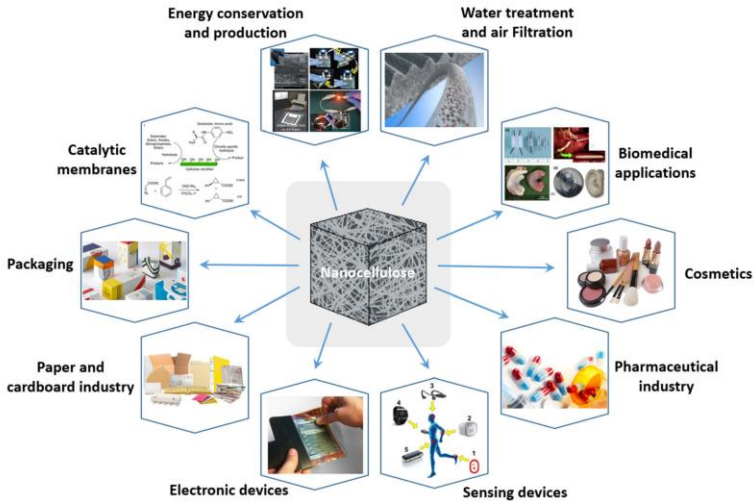

Figure 6: State of the art of applications for new nanocellulose-based materials [90]

Cellulose fiber composites have great potential in the automotive industry, where lightweight, low-cost, durable, and recyclable parts are needed. Natural fibers meet these requirements, and they are also able to increase stiffness and improve sound absorption. In 2002, the use of such composites in the automotive industry increased by 54% per year, especially in interiors [69].

Smart nanocellulose-based materials can respond to light, temperature, electrical impulse, pH, and magnetic force stimuli.

With its biocompatibility and biodegradability, nanocellulose is a suitable material for electro-stimulated drug delivery systems. It has also been widely used as a reinforcement in shape memory materials that change shape based on an external stimulus. Due to its low density, chemical and biological resistance, thermal stability, and high mechanical strength, it can be used as a functional material in actuation systems and sensors for detecting pH, organic vapors, ions, and humidity. Another possibility is the application in OLED displays or paper transistors [1].

Stability up to 350 °C makes nanocellulose a good insulation material. Miao and Hamad [69] also add high noise absorption performance due to the hollow structure of the fibers and the relatively reactive surface of cellulose. Hydrogen bonds between cellulose chains can form a uniform layer on a hydrophilic substrate. Viscosity decreases with increasing shear rate. With these properties, nanocellulose is suitable for paints, and coatings, as a rheological modifier for foods, as well as in cosmetic and pharmaceutical products [34].

Nanocellulose in the form of a hydrogel is capable of inducing wound healing on its own without any additives. CNFs are used as a wound dressing due to their ability to form translucent films and absorb fluids. Such a dressing can maintain the appropriate moisture for the type of wound. The transparency of nanocellulose also makes it easier to monitor the development of the wound without having to remove the dressing [10].

Nanocellulose can also be added to adhesives and coatings. In agglomerated wood materials, adhesives play an important role and the addition of nanocellulose improves both its mechanical and physical properties and increases its quality. Veigel et al. [70] investigated the effect of CNFs on the properties of urea-formaldehyde (UF) and melamine-urea-formaldehyde (MUF) adhesives. Particleboard with UF adhesive and CNFs content of 1 wt.% showed less thickness swelling and better joint and bending strength than when using pure UF adhesive. The reinforcing effect of CNFs was even more pronounced in OSB boards, where the strength increased by 16%. The reinforcing effects of CNFs in adhesives have been studied by several authors and their results

were summarized by Vineeth et al. [71] in their review. Chaabouni and Boufi [72] added 1–10 wt.% CNFs to polyvinyl acetate adhesive. With the increasing addition of nanocellulose, the viscosity of the adhesive increased, and the shear strength of the bond improved both in dry and wet conditions and in the case of 10 wt.%, approximately twice as much as in pure adhesive. Furthermore, improvements in water resistance and wet mechanical properties were observed at CNF contents above 7 wt.%.

Another application is nano-coatings. Nanoparticles can interact with their surroundings due to their morphology, and their nanoscale dimensions ensure that transparency is maintained. Nanocoating is a process in which a layer with a thickness of less than 100 nm is applied to the treated material, the task of which is mainly to improve durability, mechanical properties, frost resistance, UV radiation adsorption, and reduce water absorption. The advantage of coating application is that it is applied to the finished product. When using nanocellulose coatings on paper, oxygen barrier and oil resistance can be increased. At the same time, high print quality, water resistance, and dimensional stability can also be achieved [74]. Veigel et al. [93] added CNFs to an oil-based coating applied to oak wood. The result was an improvement in abrasion resistance and, to a lesser extent, chemical resistance. According to them, the improvement in resistance was due to the CNFs network, which prevented the oil from penetrating the wood and thus formed a protective CNFs/oil composite layer on the surface.

In addition, nanocellulose has been recognized for its potential in conservation efforts. Its application in the restoration of historical documents demonstrates its ability to provide excellent stability against aging and environmental degradation, making it a valuable material for preserving cultural heritage [53].

Biopolymers usually have poorer mechanical properties than synthetic polymers. Therefore, nanocomposites have been developed. Due to their natural biocompatibility, renewability, biodegradability, and versatility, CNFs are one of the best reinforcement materials. They can be functionalized without

disrupting the main molecular structures, which facilitates uniform dispersion and strengthens the bond to the polymer matrix.

According to Kumar et al. [33], CNFs have great potential as reinforcements and provide higher stiffness, tensile strength, thermal stability, and barrier properties in polymeric materials. The high aspect ratio helps transfer load from the matrix to the material [1]. In addition, due to their lower density, biodegradability, and excellent mechanical and thermal properties, they are a good alternative to artificial fillers. However, due to the hydrophilicity of nanocellulose, the biocomposite may degrade faster than the original composite at a temperature of 20–30 °C. Besides high moisture absorption, other disadvantages are poor wettability and incompatibility with most of the polymer matrix [33].

One of the most studied areas is using nanocellulose crystals (CNCs) as a reinforcing component of polymers. CNCs are very good nanofillers due to the abundance of hydroxyl groups, reactivity, large specific surface area, and their mechanical, thermal and optical properties. In polymers, they cause an increase in tensile strength and a decrease in elasticity, which is caused by strong intermolecular bonds such as covalent bonds, van der Waals forces, and mechanical bonds between CNCs and the polymer matrix. They also improve corrosion resistance. Compared to conventional nanofillers such as carbon black, silica, mica, etc., CNCs have a lower negative impact on health and the environment [9]. The environmental friendliness of biopolymers and biocomposites is discussed in the book by Mohanty et al. [89]. CNCs in thermosets such as some polyurethanes, unsaturated polyesters, phenolic and epoxy resins synergize with the polymer matrix, and the result is a high-quality material with excellent properties. However, compared to thermoplastics, the disadvantage is the high temperature and curing time or problems with recycling. In addition to improving strength, stiffness, and corrosion resistance, the addition of CNCs could reduce the internal stress arising during curing and increase the density, thereby supporting the improvement of toughness [9]. After adding CNCs to a composite based on styrene-butyl acrylate

copolymer, thermal stability improves. Miao and Hamad [69] found in their research that after adding 6 wt.% CNCs, the material is stable up to almost 500 K compared to samples without the addition of CNCs, whose mechanical properties were significantly reduced already at a temperature of 350 K.

Nanocellulose can also be used as an additive to the polymer matrix of natural or synthetic fibers of textile materials or it is applied to their surface. This is to improve self-cleaning, durability, water and dirt repellency, absorbency, and dyeability, as well as improve the physical and thermal properties of fabrics [33].

Petroleum-based materials are used as packaging materials due to their barrier properties, suitable gas and water content, and low price. Concerning the environment, an environmentally friendly and sustainable alternative is currently being sought for these non-degradable and non-recyclable materials. However, biopolymers do not have sufficient barrier properties. By adding nanomaterials to biopolymers used for packaging, a higher crystallinity of the material was achieved. Due to the decrease in the density of surface micropores, the barrier properties improved. Transparency and biodegradability are also positive aspects [1]. Packaging materials should protect the product from chemical, physical, and biological damage. They should therefore have a good barrier against oxygen, water vapor, grease, and microorganisms [27]. Kim et al. [1] wrote that cellulose nanowhiskers (CNWs) can reduce water permeability by 82% and oxygen permeability by 90%. Chinga-Carrasco and Syverud [74] studied nanofibril-based films in terms of barrier properties. The OTR (oxygen transmission rate) value recommended for packaging is below 20 mL/m^2.day.atm. The lowest value measured in their study was 3 mL/m^2.day.atm, with values ranging from 3–4.4 mL/m^2.day.atm. Therefore, it can be seen that these values are much lower than recommended. However, Silva et al. [75] in their scientific paper on packaging materials mention that the nanocellulose coatings improved mechanical strength, grease resistance, and reduced oxygen permeability, while water vapor permeability was still high. They found that although nanocellulose helps to bring biocomposites closer to packaging requirements, oxygen, and

water vapor permeability is still higher than that of petroleum-based plastics, and therefore further improvements to these composites are needed. Packaging that has high barrier properties improves food protection during transportation and storage and increases its shelf life [36]. European Regulation (EC) No 10/2011 recognizes cellulose and some of its derivatives as safe for food packaging, use in polymers, etc. However, studies on the toxicity of nanocellulose show mixed results, and its negative effects have not been completely ruled out [75].

7. Use of Nanocellulose in the Pulp and Paper Industry

One of the applications of nanocellulose that is being investigated is its use in the paper-making industry. This involves adding renewable, environmentally friendly "green" nano additives to enhance paper properties or create papers with specific properties. This can improve mechanical strength and physical and printing properties, but also recyclability. The unique properties of nanocellulose (large specific surface area, good optical properties, low weight, high stiffness, and strength) and the properties derived from cellulose (biodegradability, renewability, and sustainability) make nanocellulose an excellent additive for papers. Nanocellulose has also been added to special papers, namely waterproof papers, so-called "fresh" papers, insulating papers, technical papers, anti-stain papers, or packaging papers [34,32].

Paper can be simply described as a network of interconnected natural cellulose-based fibers, whose tensile strength is close to that of hardened steel. However, the highest tensile strength measured for paper was only about a quarter of the strength of individual fibers. During drying, hydrogen bonds form between the fibers. The strength of paper depends on both the strength of these hydrogen bonds and the strength of the individual fibers. Covalent bonds are stronger in cellulose molecules, while hydrogen bonds between fibers are much weaker. The relationship between bond and fiber strength is very important. In addition, the strength of the paper is affected by the length of the fibers, the contact area between the fibers, and the orientation and distribution of the fibers [76].

Nanocellulose fibrils (CNFs) can be mixed directly into the pulp with or without retention agents or can be pre-coated with retention agents on a feedstock component such as a binder or a fibrous fraction. It is therefore possible to control whether the CNFs will be present freely or unbound in the mixture before dewatering or whether they will be coated as a surface layer on one or more fractions using retention agents. The surface of CNFs is rich with free hydroxyl groups. When added to the pulp, they will be distributed in the gaps between the primary fibers or on their surface and closely combined with the pulp fibers (Figure 7), which strengthen the adhesion between the fibers, fill the voids in the paper, and achieve the effect of improving the strength of the paper [77].

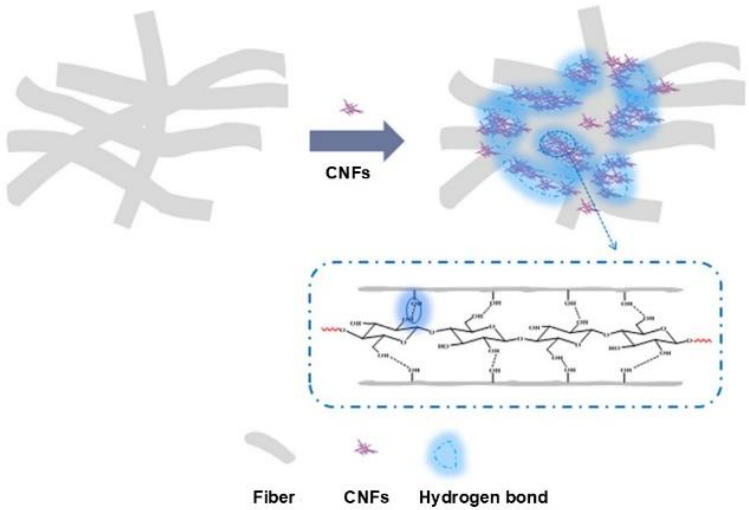

Figure 7: The combined effect of fiber entangling and NC reinforcement within the paper [34]

Coating materials containing CNFs can be applied to paper using conventional techniques such as bar, press, or roller coating, but spray or foam coating can also be used. However, the application of CNF suspensions is limited by their high viscosity. It has been found that coatings containing CNFs reduce the surface strength of coated paper compared to coatings containing only latex, therefore offset printing is not suitable. At the same time, inkjet printing is also not very suitable because this type of coating

reduces its speed and increases its density. Even so, CNF coatings are generally used to improve the surface or certain properties of paper, e.g. in the case of newsprint for reducing the tendency of sheets to pill. A hydrophobic film is formed on the surface of the paper, which increases water resistance, surface strength, and bending resistance. The quality of the coating is closely related to the number of layers and their thickness [78].

Nanocellulose as an additive is environmentally friendly and renewable, but it also has a high adsorption capacity and limited single-use effect. Some researchers have responded to this by using cationic polymers as additives to nanocellulose, which should result in a synergistic effect of these components. Also, the addition of carboxymethylated or TEMPO-oxidized CNFs to the primary fibers caused an improvement in the mechanical, chemical, and optical properties of paper [34].

The optical properties of paper can be significantly affected by the addition of CNFs. The density of the sheet increases, the binding potential increases, and at the same time the specific surface area and the light scattering coefficient decrease. As a result, the brightness and opacity are reduced. In more fibrillated CNFs, the light scattering coefficient was lower because the density and the bonded area increased more than in less fibrillated CNFs [78].

Other properties are also affected by the addition of nanocellulose. The smoothness of the paper increases and the permeability to air and other gases, and even oils, decreases due to the closure of open pores. The paper with a double CNF/CNC coating (Figure 8) had about 300 times less air permeability, 260 times less oxygen permeability, and a 30% lower water vapor transmission rate. The paper thus treated is therefore an ideal packaging material resistant to gases and oils [34].

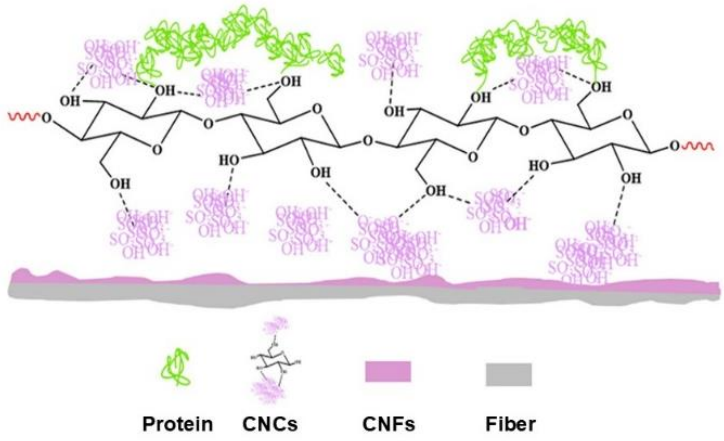

Figure 8: The enhanced adhesion of multilayered CNF and CNC coatings [34]

Paper additives, besides basic fillers and binders, are most often various auxiliary substances, retention agents, sizing agents, dyes, optical brighteners, substances for increasing strength in the dry and wet state, defoamers, dewatering agents, wetting agents, flocculants, coagulants, dispersants, etc. [87,88]. Nanocellulose, as a paper additive, has at least retention, drainage, and strengthening effects. Retention agents, important mainly in the drainage phase, play an important role, especially in recycling. Nanocellulose could improve the physical properties of paper by creating surface contact bonds or better connecting the fibers with the filler. A small dose of retention agent does not have such a high retention effect, on the other hand, a large dose causes the formation of large flakes, which affects the uniformity of the paper. Therefore, two-component agents have been developed, which should be more effective. Nanocellulose could also be a suitable additive to papers produced from waste paper, thereby increasing the utilization rate of recycled paper. Dewatering agents are intended to improve the drainage and dehydration rate of wet paper. At the expense of reducing the surface tension and fiber contact angle, the wettability of the surface deteriorates and drainage problems occur. Since CNFs fill the open pores, the drainage time is extended. To eliminate the negative effect, it is possible to use only a small addition of CNFs with suitable retention additives. To control chemical flocculation, and

optimize retention and drainage, dual retention systems based on a combination of polymers (e.g. cationic polyacrylamide - CPAM, chitosan) and CNFs microparticles have been developed. However, nanocellulose is itself a material capable of adsorption, which gives it the prerequisites to be a dewatering agent. It can be modified to improve its affinity and adsorption capacity for specific substances [34]. Modification with nanocellulose could lead to an increase in binding capacity, it would be possible to control binding interactions with fibers and fillers, and reduce the consumption of retention agents [32].

If the aim is to increase the strength of the final product, nanocellulose is usually added to the pulp. However, the resulting hydrogen bonds could increase the cost of effective dispersion and drainage and reduce brightness. Some of these problems can be overcome by paper treatment with a nanocellulose coating. However, in this case, problems arise with its homogeneous distribution and adhesion to the paper. Other difficulties are caused by the size and high swelling of nanocellulose. Due to the reduction in porosity, the drainage time is extended, which reduces productivity, but the resulting paper has excellent barrier properties [32].

The recommended effective amount of nanocellulose addition is 2.5–10%, but to avoid drainage problems, lower amounts are used in practice [78].

Nanocellulose-based nano papers are highly transparent, biodegradable, have high thermal stability, functionality as well as printability, and can be used in electronic devices such as sensors, actuators, etc. [33].

Many uses of nanocellulose have been discussed, e.g. as a retention agent, wet strength aid, flexographic ink removal, pilling control, or for specialty papers requiring antibacterial, flame retardant, or electrical properties. However, due to the high production capacity, nanocellulose has not yet been fully commercialized in industrial production.

8. The Influence of Nanocellulose on Paper Strength Characteristics

Kim et al. [1] compared nanocellulose with other materials such as glass fibers, Kevlar, graphite, or steel wires and found that CNCs (cellulose nanocrystals) and CNTs (cellulose nanotubes) are extremely strong, which is evident from Table 2. Hashemzehi et al. [36] mention that the tensile strength of CNFs is almost twice as high as, for example, carbon steels.

Table 2: Physical-mechanical properties of various materials compared to anocellulose [3]

Material	Tensile strength (GPa)	Young's modulus (GPa)	Density (g/cm^3)	Tensile / density	Modulus / density	Thermal exp. coe. (ppm/K)
CNCs	7.5	145	1.6	4.7	90.6	3–22
Glass fiber	4.8	86	2.5	1.9	34.4	13
Steel wire	4.1	207	7.8	0.5	26.5	15
Kevlar	3.8	130	1.4	2.7	92.9	-4
Graphite	21	410	2.2	9.5	186	2–6
CNTs	11–73	270–970	1.0	11–73	270–970	-

The mechanical properties of nanocellulose depend on the source from which the nanocellulose is obtained, as shown in Table 3. However, this factor is also influenced by the molecular level, shape, and quality of the fiber. There can be variability between the properties of nanocellulose obtained from the same source, but in different plant growth environments or with various ratios of cellulose, hemicelluloses, and lignin. The fiber aspect ratio and the mechanical stiffness are influenced by the production technology. A higher aspect ratio of the filler materials results in improved stiffness and stress transfer and deterioration of dispersion uniformity in the matrix. The stiffness of cellulose depends on its crystallinity and molecular arrangement, while the strength of the fibers increases linearly with increasing cellulose content and decreasing microfibrillar angle. The strength of the fibers is not greatly affected by the structural arrangement, but rather by internal structural defects [1].

Table 3 Comparison of mechanical properties of natural fibers [79,1]

Fiber	Tensile strength (MPa)	Elongation at break (%)	Young's modulus (MPa)
Cotton	264–654	3–7	4980–10,920
Wool	120–174	25–35	2340–3420
Silk	252–528	20–25	7320–11,220
Flex	300–900	2.7–3.2	24,000
Jute	342–672	1.7–1.8	43,800
Sisal	444–552	2–2.5	-
Ramie	348–816	3.6–3.8	53,400

As mentioned earlier, the addition of CNFs increases the stiffness and strength of biocomposites. However, the orientation of CNFs is also an important factor. The longitudinal orientation is the best in terms of mechanical properties, while the transverse orientation is the least desirable in this regard. Joseph et al. [79] investigated the effect of fiber orientation on tensile strength and Young's modulus of PP/sisal composites. Their results can be readily observed from the graphs in Figure 9, from which these trends are evident.

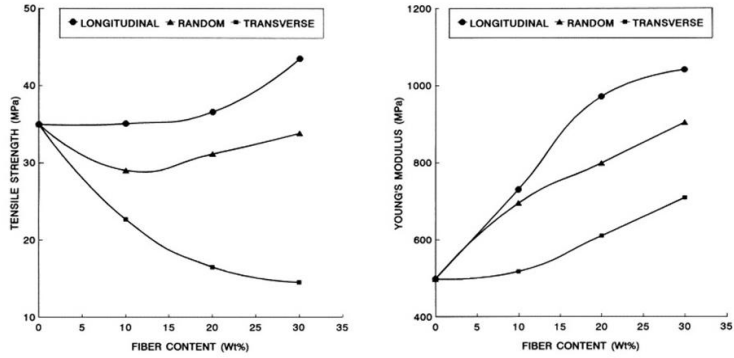

Figure 9: Effect of fiber orientation against fiber content on tensile strength and Young's modulus of PP/sisal composites [79]

CNFs in paper increase mechanical strength by increasing the bonding surface area. They act as fillers in the pores between the fibers, filling the voids and forming hydrogen bonds between fibers as shown at the beginning in Figure 1.

The strength of paper is determined by three components: the strength of the fibers themselves, the strength of the bonds between the fibers, and the degree of fiber cross-linking. The large specific surface area gives nanocellulose a high potential for hydrogen bonding. Hydroxyl (–OH) groups on nanocellulose can form stronger hydrogen bonds with hydroxyl groups on the primary fibers of the reinforced paper, thereby increasing mechanical strength. The reinforcing effects of nanocellulose may lie in its functioning as an adhesion promoter, helping to glue the fibers together and increasing the contact area. Another possibility is that nanocellulose creates crosslinked structures between the fibers, thereby increasing the load-bearing capacity of the fiber. The amount of additive used is important. Although it has been proven that nanocellulose improves the properties of the matrix, when added in excessive amounts, it can have a negative effect [34].

Hashemzehi et al. [36] reported that coating the paper by CNF does not affect tensile strength, but significantly improves bending stiffness.

A parameter that also affects the fiber's mechanical strength is the degree of fibrillation (Figure 10). Wider particles do not cause such an improvement in mechanical properties, but more effective retention. A higher degree of fibrillation should facilitate the formation of a homogeneous structure and thus improve strength. On the other hand, a smaller length and aspect ratio do not allow the fibrils to strengthen the interfiber bonds to a greater extent [36]. The results published by Syverud et al. [80] show that with higher fibrillation, the density increases but the tensile strength decreases. According to them, this was probably caused by too pronounced fibrillation, which shortened the fibers.

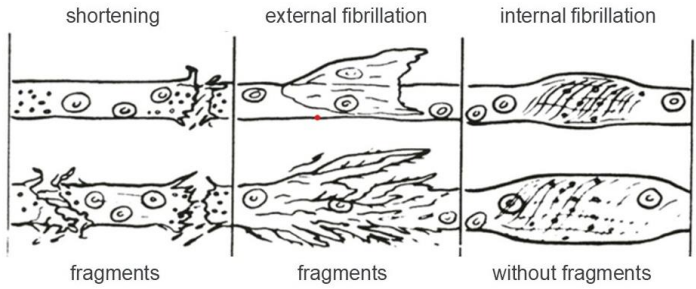

Figure 10: Types of fibrilation [87,88]

Strength can also be increased by adding water-soluble hydrophilic dry strength additives such as starch, which also can improve retention. Cationic starch acts as a fixative, trapping nanofibers in the cellulose network of primary fibers [81].

In a study by Syverud et al. [80], the tensile strength of thermo-mechanical pulp sheets increased by more than 20% due to the addition of 4% microfibrillar cellulose (MFC). Taipale et al. [81] reported a linear increase in tensile strength with increasing MCF content. According to them, the increase in the number of bonds prevents fiber slippage and thus increases strength. The addition of 100 mg/g MFC caused a 17% increase in tensile strength and the addition of 45 mg/g MFC caused a 57% increase in out-of-plane strength. They mention that at lower MCF additions, the out-of-plane strength shows a similar trend to the tensile strength, but at higher additions, the increased proportion of fibrils may introduce delamination planes into the structure, which act as low-energy fracture paths and limit the increase in out-of-plane strength.

Tozluoglu and Poyraz [82] investigated the differences between kraft pulp and $NaBH_4$-modified pulp. In both types, the tensile strength increased with the addition of cellulose microfibrils (CMF) and cellulose nanofibrils (CNF). 1.5% CNF addition increased the tensile strength of kraft pulp by 5.48–15% and of $NaBH_4$-modified pulp by 0.93–5.59%. Higher additions of CMF/CNF caused further increases. The highest values of 19.5% and 14% were found for the addition of 3% CMF.

Žďárský [83] evaluated the increase in the strength characteristics of sheets produced from three types of bleached and unbleached kraft pulp after adding 0.5%, 1%, and 3% nanocellulose in the form of a hydrogel. The results of this work showed an increase in the tensile strength with increasing addition of CNFs. Lower values were found for bleached kraft pulp, where the highest increase of 13.5% was found with the addition of 0.5% nanocellulose. The tensile strength was increased the most with the addition of 3% nanocellulose to unbleached pulp, namely by 61–66%. The same trend was also observed in the case of ductility. However, the highest increase of up to 123% was found in bleached kraft pulp samples with the addition of 3%. The increase of ductility was not as great for unbleached pulp. The breaking length also increased with increasing CNF content - the greatest increase of 63 % was observed at unbleached kraft pulp with a 3% addition of CNFs. In most cases, the strength characteristics of unbleached kraft pulp were higher than those of bleached kraft pulp.

Lošťáková [84] assessed the effect of CNFs on the strength properties of an industrially produced fiber composite material used in the automotive industry, where high strength is required. The preparatory study consisted of testing the effect of adding CNFs only to primary cotton fibers, which were only pulped in one series and pulped and refined in the second series. Subsequently, sheets of fiber composite material were produced from pulped and pulped and refined cotton. CNFs were added to all batches in amounts of 0% (reference samples), 0.5%, 1%, and 3%. The measured parameters were: thickness, porosity or air permeability, and tensile index. The results of the measurements confirmed the expected trends, namely an increase in density and tensile index. However, the porosity decreased significantly. The most optimal results for the fiber composite material were observed using refined cotton with CNF content of 1%, which caused the increase of dry and wet tensile index by 24.6% and 21.9%, respectively, while the porosity decreased by 29.4%. In general, the tensile index values were higher for pulped and consequently refined cotton sheets, but the effect of nanocellulose on the pulped cotton sheets was greater.

On the contrary, Perdoch et al. [85] presented the results of a study where they looked at the influence of different nanocellulose structures on the strength properties of specially treated Whatman absorbent paper, and in their case, the addition of CNF(Et) even reduced the tensile strength of the paper by 40% compared to untreated paper. CNCs fared better, with increased tensile strength and tear resistance. They cite the lower dipole moment of ethanol compared to water as the reason for the strength reduction with CNF treatment.

Therefore, it is obvious that more laboratory-scale research studies are needed in order to improve the targeted use of nanocellulose for each special application. However, the nanocellulose market continues to expand regardless, and various producers and research institutions actively developing new processes and applications to different end-user utilization.

Ciriminna [94] noted that the main investors in nanocellulose research and production are primarily from the forest and paper industries, indicating a strong interest in integrating nanocellulose into existing production lines. However, challenges remain in scaling up production and reducing costs to compete with conventional materials.

9. Conclusion

This book chapter reviews the recent advancement of nanocellulose production from cellulose, summarizes its properties, and discusses its possibility for sustainable future materials focusing on the utilization in the paper and packaging industry in the form of environmentally friendly fiber composite materials that surpass current technological limits of existing products.

Overall, the incorporation of nanocellulose fibers into paper products not only enhances their mechanical and optical properties but also plays a crucial role in promoting sustainability within the paper sector. By utilizing renewable resources and enhancing the recyclability of paper products, nanocellulose contributes to a more circular economy.

As research continues to explore the full potential of nanocellulose, its role in the paper industry is likely to expand, paving the way for innovative applications and improved product performance.

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