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Production of nanocellulose from lignocellulosic biomass and its potential applications: A review

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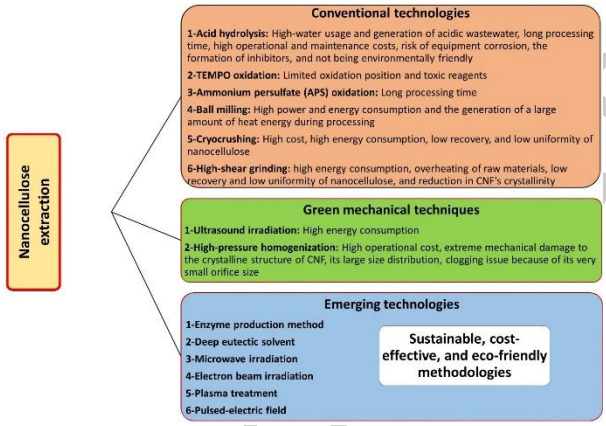
Graphical abstract



Abstract

Lignocellulosic biomass is a complex natural polymer primarily composed of cellulose, hemicellulose, lignin, and various other chemical components. The cellulose in the lignocellulosic biomass can depolymerize into a nano-dimension biomaterial called nanocellulose, which possesses unique features with potential application in various fields. Nanocellulose production from lignocellulosic biomass has become the subject of extensive research in the last few decades in the fields of material sciences and biomedical engineering and has attracted the attention of scientists and technologists worldwide. This production faces many challenges in utilizing the cellulose from lignocellulosic biomasses and subsequent processing for their conversion into nanocellulosic materials and their further applications in various fields of science and technology. This current review not only focuses on the production of nanocellulose from lignocellulosic biomass through different production methods but also discusses various sources, types, properties, and their applications in material science and biomedical engineering. This research review certainly shows that in the future, nanocellulose has great potential to be used as a renewable source in the field of sustainable materials and nanocomposites.

Keywords: Lignocellulosic biomass, cellulose, nanocellulose, production and applications.

1. Introduction

One of the significant challenges of this century is the synthesis of new materials that fulfill our needs and, at the same time, are eco-friendly and renewable. This goal has led to enormous growth in research, especially in the field of bio-based polymeric materials and composites during the last two decades (Gejo *et al*. 2010; Pandey *et al*. 2010). Compared to synthetic materials, bio-based materials are easily decomposable, regenerated naturally, and have no adverse effects on the environment (Tabone *et al*. 2010; Yates and Barlow 2013). These biopolymers are abundant in nature with numerous applications potential in material sciences (Ditta 2012; Ditta and Arshad 2016; Mehmood *et al*. 2023; Mehmood *et al*. 2023; Mehmood *et al*. 2022). Among various biopolymers, cellulose has been frequently chosen, as it is the most abundantly present biomaterial on earth with less cost, is eco-friendly, and is renewable (Azizi *et al*. 2005)

The cellulose from a natural source contains both crystalline (proper arrangement of molecules) as well as amorphous (lack of arrangement of molecules) structures. The extent of crystallinity and amorphous structure mainly depends on the source from which the corresponding cellulose is extracted, purity, time, and

temperature. (Revol *et al*. 1994). When cellulose molecules are subjected to a set of experimental conditions like mechanical, chemical, and enzyme actions, it leads to the production of crystalline structures called cellulose nanocrystals (CNCs) (Domingues *et al*. 2014). The bulk form of cellulose has higher amorphous fractions than CNCs. The cellulose nanocrystals are composed of rod-shaped molecules in a proper arrangement, which builds up the crystalline structure. The synonym terminologies used for CNCs are whiskers, nanocellulose, nanofibers, and microcrystalline, but CNCs are the most appropriate and widely accepted nomenclature used (Habibi *et al*. 2010). The crystalline nature of CNCs is responsible for a specific strength, biodegradability, useful rheological behavior, modulus, high surface area, maximum chances of chemical modification, biocompatibility, lightweight, high aspect ratio, outstanding oxygen permeation resistance, oil grease barrier, water vapor transmission rate resistance, aqueous liquid barrier, and unique liquid crystalline properties. (Islam *et al*. 2013; George and Sabapathi 2015; Imani *et al*. 2022). Many researchers have produced CNCs from various cellulosic sources e.g. durian rind, wheat bran, corn stover, sugarcane bagasse, sago seed shells, raw cotton linter, sisal fibers, mengkuang leaves, kusha grass, mulberry, cotton stalks, and many others (Li *et al*. 2009; Sheltami *et al*. 2012; Morais *et al*. 2013; Nguyen *et al*. 2013; Penjumras *et al*. 2014; Wang and Zhang 2014; Costa 2015; Soni *et al*. 2015; Naduparambath *et al*. 2018; Rehman *et al*. 2023; Rehman *et al*. 2023; Khan *et al*. 2023). In comparison to traditional composite materials, CNCs exhibit high mechanical, electrical, optical, thermal, and gas barrier properties (Rehman *et al*. 2023, Le Bras etal. 2015; Guidetti *et al*. 2016; Leonardo *et al*. 2016; Shishehbor and Zavattieri 2019). Therefore, the global scientific community is focusing on the use of CNCs for various industrial applications like packaging, automobile, thermal management, and optical industries (Khan *et al*. 2023, Siqueira *et al*. 2010; Duran *et al*. 2012; Charreau *et al*. 2013). Due to their exceptional properties, CNCs have been used as a reinforcement component in nanocomposites (Liu *et al*. 2015; Yoo *et al*. 2017), barriers in packaging applications (Lavoine *et al*. 2012), transparent media in organic electronics, and anti-counterfeiting in security applications (Chindawong and Johannsmann 2014). The CNCs can also be utilized in film, aerogel, and foam, depending on the required application (Khan *et al*. 2023)

CNCs have been used as ideal nanofillers to improve the strength and modulus in the field of polymer nanocomposites. These have been used in both synthetic and natural polymers for the development of nanocomposites. Many synthetic polymers such as polycaprolactone, polyethylene, polyvinyl chloride, polyvinyl alcohol, polyurethane, and polypropylene have been used in the preparation of nanocomposites (George and Sabapathi 2015; Ljungberg *et al*. 2005; Cao *et al*. 2007; Goffin *et al*. 2011). Similarly, numerous naturally available polymers such as chitosan, natural rubber, starch, hydroxypropyl methylcellulose, gelatin, cellulose acetate butyrate, soy protein, and carboxymethyl cellulose have also been used in this regard (George and Sabapathi 2015; Choi and Simonsen 2006; Wang *et al*. 2006; Bendahou *et al*. 2009; de Mesquita *et al*. 2010).

Another distinctive application of CNCs is their use in coatings (Khan *et al*. 2022). Due to their high stiffness, strength, and barrier properties, these offer enhanced mechanical, scratch-resistant, gas/water barrier, and weathering-resistant properties to the coatings. The traditional coatings are mostly oil- or solvent-based, which release volatile organic compounds (VOCs), thereby polluting the environment. The use of CNCs in different coatings such as waterborne coatings is environmentally friendly and therefore, has become the focus of intense research for scientists and technologists worldwide. Based on the above discussion, CNCs have great potential to be used in the field of coatings due to their state-of-the-art properties. The present review summarizes the conversion of lignocellulosic biomass into nanocellulose, its production methods, and its potential applications. In addition, it also highlights the main conclusions made and future research directions by finding research gaps.

2. Lignocellulosic biomass

Lignocellulosic biomass is an abundantly available complex biopolymer composed of cellulose, hemicellulose, and lignin with a small number of inorganic materials and extractives (Demirba 2005; Li *et al*. 2015). These polymeric constituents are related to each other in a heater matrix to different degrees and varying compositions, depending on the species and even the source of the biomass from a species (Himmel *et al*. 2007). The worldwide annual production of lignocellulosic biomass is approximately 1.815×10^{11} metric tons Paul and Dutta 2018). Due to its higher availability and renewable nature, it has been one of the world's most potential resources having diverse applications. However, various pre-treatment steps are required for the successful separation of the cellulosic portion from tightly bound polymeric constituents in lignocellulosic biomass. The presence of cellulose in lignocellulosic biomass is capable of depolymerization into a biomaterial with nano-dimensions, excellent mechanical properties, rich in hydroxyl groups available for modification, high surface area, and natural features with eco-friendly applications in the biomedical field, reinforcement of nanocomposite material, pharmaceutical carriers, electronic substrates, and many others (Lee *et al*. 2014).

2.1. Cellulose

Cellulose is one of the natural, renewable, and biodegradable polymers that is abundantly available on Earth (Mehmood *et al*. 2023; Mathew *et al*. 2006; Nada *et al*. 2009). It is an essential structural component of the cell wall present in many plants, including reeds, grasses, and woody vegetation. In addition to plants, cellulose also occurs in many living organisms such as fungi, algae, bacteria, and even in some marine animals. It is estimated that $\sim 1.5 \times 10^{12}$ tons of cellulose are synthesized each year around the globe (Klemm *et al*. 2005). Cellulose is a natural linear polymer (polysaccharide), composed of β-D-

glucopyranose units connected via β-1-4-linkages (Brinchi *et al*. 2013). The cellulose present in nature does not exist in the form of an isolated individual molecule, but it occurs as assemblies of individual cellulose chain-forming fibers. These fibrils are combined into bigger units named microfibrils, which are subsequently assembled into a fiber. These microfibrils contain both crystalline (highly ordered) as well as amorphous (lack of ordered) regions. In the crystalline areas, cellulose chains are strongly packed together by strong inter and two intramolecular hydrogen bonds (Zhou and Wu 2012).

Common sources of cellulose include cotton linters, jute, and wood pulp. Cotton linters contain approximately 90-95% cellulose, while in jute and wood pulp, it is around 45-63 and 40-45%, respectively (Pickering 2008). Cellulose extracted from highly pure cotton linters requires minimal treatment (NaOH treatment) to remove non-cellulosic materials. However, other fibers such as jute, wood pulp, hemp, and ramie need extensive processing to remove the non-cellulosic components. Cellulose extraction from lignocellulosic biomass is a topic of intense research with the advent and advancement of nanotechnology. It attracts further interest in the new form of "nanocellulose" to be used as a novel and advanced material and has become the principal theme of research work carried out by scientists and technologists worldwide (Chirayil *et al*. 2014).

2.2. Hemicellulose

Hemicelluloses are heterogeneous polysaccharides present in the cell walls of almost all plants and act as a linkage molecule between cellulose and lignin. Hemicellulose polysaccharide chains are relatively short with an average degree of polymerization of about 200. Hence, these can easily be hydrolyzed and extracted from plant tissue by using water, acid, or aqueous alkali. Unlike cellulose, it is an amorphous polymer with little strength (Chirayil *et al*. 2014; Mäki-Arvela *et al*. 2007). Hemicelluloses like many other polysaccharides, act as substantial barriers to oxygen. However, these are moisture-reactive and show low barrier resistance under humid conditions. Mannan-type hemicelluloses have successfully been used in the preparation of packaging films, showing strong film-forming properties and resistance to oxygen diffusion (Aulin and Ström 2013).

2.3. Lignin

Lignin, the second-most abundantly present natural polymer on earth, binds with cellulose and hemicellulose and is mainly responsible for supporting plant structure. It primarily consists of p-hydroxyphenyl, guaiacyl, and syringyl units (Huang *et al*. 2019). It is composed of complex aromatic polymers, which are found in different proportions among various cellulosic biomasses. It has been wonderful raw biomass, being extensively used in material science due to the presence of various functional groups, degradability, renewability, low cost, and non-toxic nature (Liu 2010; Zhang *et al*. 2021).

2.4. Additional chemical components

Besides cellulose, hemicellulose, and lignin, cellulosic biomass also contains pentosans, extractives, and inorganic materials. These extractives are small molecular-weight organic substances that exist in the form of monomers, dimers, or polymers in the cell wall (Rowe 2012). These chemicals are comprised of fats, fatty acids, steroids, fatty alcohols, terpenes, phenols, resin acids, rosin waxes, and many other minor organic compounds. In softwoods, the contents of extractives are higher than in hardwoods (Siqueira *et al*. 2010).

3. Main sources of cellulose

Plants are the major sources of cellulose, but bacteria, fungi, and other marine animals can also produce cellulose in a significant amount (George and Sabapathi 2015; Gallo Stampino *et al*. 2021). A brief description of the significant cellulose sources is given below:

3.1. Plants

Plants are the primary potential source of cellulose due to their natural abundance. In plant sources, wood pulp and cotton fibers are the primary sources of cellulose and can be processed to extract cellulose on a commercial scale (Dalle Vacche *et al*. 2021; Mehanny *et al*. 2021). A large quantity of cellulose can also be obtained from other plants such as grasses, aquatic plants, ramie, jute, sisal, hemp, flax, etc (Mehmood *et al*. 2023; Anh *et al*. 2021). Agricultural wastes such as rice and wheat straw, sawdust, cotton stubbles, sugarcane bagasse, etc. are also among the significant sources of cellulose (George and Sabapathi 2015; Mateo *et al*. 2021; Maslennikov *et al*. 2022).

3.2. Algae

Cellulose is the fundamental component of the cell wall of many algal species. Among algal species, blue-green algae is a major source of cellulose while red and yellow algal species contain smaller amounts of cellulose. The cellulose extracted from valonia and cladophora species is crystalline (95%). The properties of microfibrils depend on the nature of the cellulose biosynthesis process occurring among different algal species (Park *et al*. 2010).

3.3. Bacteria

Komagataeibacter xylinus is a well-known bacterial species that produces cellulose using nitrogen and carbon to create dense and clear cellulose microfibrils (Paramasivan *et al*. 2022). The cellulose extracted from microbes is unique possessing outstanding properties due to its nanostructure, purity, and good mechanical strength compared to the cellulose derived from plants (Klemm *et al*. 2005).

3.4. Tunicates

Tunicates, belonging to marine invertebrates families, can also produce large quantities of cellulose. Cellulose production is mainly dependent on the enzyme complexes present in the epidermal layer of tunicates. The properties of cellulose microfibrils vary among species and production processes (Habibi *et al*. 2010; Chirayil *et al*. 2014; Jonoobi *et al*. 2015).

4. Nanocellulose

Nanocellulose is a remarkable natural fiber, isolated from cellulose due to its small size, generally 100 nm or less in

diameter and some micrometers in length. It is a biodegradable nanoparticle with excellent strength, lightweight, and low density. It has many important features that make it an outstanding material for biomedical applications, nanocomposites, paints, coatings, adhesives, energy, environment, and many others (Mondal 2017; Pirozzi *et al*. 2021). The stiffness of nanocellulose is very high, and it is up to 220 GPa of elastic modulus, which is higher than Kevlar fiber. It has an excellent tensile strength value of up to 10 GPa which is superior to the value of cast iron. It also has a much higher ratio of strength to weight than stainless steel. It is rich in very reactive hydroxyl groups on its surface that could be modified to give different surface properties (Mehmood *et al*. 2023; Khalil *et al*. 2012; Dufresne 2013). Because of its biodegradable and renewable nature, mechanical strength, crystallinity, high specific surface area, tailorable surface chemistry, anisotropic shape, biocompatibility, and optical properties, nanocellulose has extensively been used in various scientific disciplines such as material sciences and biomedical engineering (Abitbol *et al*. 2016; Phanthong *et al*. 2018). Moreover, nanocellulose has some other important features, which have been extensively applied in the fields of packaging, filtration, electronic sensors, hydrogel, reinforcement of nanocomposites, paper and pulp, paints, coatings, and many others (Sharma *et al*. 2019).

4.1. Types of nanocellulose

Based on the technique used to synthesize nanocellulose, it is classified into three different categories, i.e. (i) cellulose nanofibers (CNFs), (ii) cellulose nanocrystals (CNCs), and (iii) bacterial nanocellulose (BNC). The technique and synthesis conditions of nanocellulose are also responsible for its dimensions, composition, and properties (Nasir *et al*. 2017).

4.1.1. Cellulose nanofibrils (CNFs)

Cellulose nanofibrils (CNFs) are cellulosic nanomaterials, which consist of linear fibrils having a diameter ranging from 5 to 50 nm and a length of a few micrometers (Lin and Dufresne 2014; Oyekanmi *et al*. 2021). CNFs comprise both the amorphous and crystalline regions in a single fiber (Jiang and Hsieh 2013) and can be extracted from cellulosic fibers by using three different kinds of processes such as mechanical treatments, chemical treatments, and a combination of mechanical and chemical treatments (Khalil *et al*. 2012). In mechanical treatment, CNFs are synthesized through high-pressure grinding of cellulosic pulp suspension, hence strong entangled networks of nanofibrils are formed (Tonoli *et al*. 2012).

4.1.2. Cellulose nanocrystals (CNCs)

Cellulose nanocrystals (CNCs) are cellulose nanomaterial with a length of 200–500 nm and a diameter of 3–35 nm. These are produced through acid hydrolysis of cellulosic substances (Samyn 2022). In this method, concentrated sulfuric acid is used to dissolve the amorphous cellulosic material into crystalline (Lu and Hsieh 2012). Hence, rod-like CNCs with nearly perfect crystallinity and almost 90% purity are obtained.

4.1.3. Bacterial nanocellulose (BNC)

Bacterial nanocellulose (BNC) is microbial cellulose produced by bacteria e.g. Acetobacter xylinum. Bacterial cellulose is different from plant cellulose as it exists as a separate molecule and further treatment to remove impurities such as pectin, lignin, and hemicellulose is not required (Lin *et al*. 2013). It has many advantages over plant cellulose, as it is highly pure with excellent permeability to liquid and gases, porosity, high water uptake, and excellent mechanical properties (Portela *et al*. 2019).

4.2. Properties of nanocellulose

The properties of nanocellulose from the plant sources mainly depend on the origin of the plant, maturity of fibers, chemical composition, defects in fibers, conditions under which the plant was grown, and the overall method used for the production of nanocellulose (George and Sabapathi 2015; Thomas *et al*. 2017; Randhawa *et al*. 2022). The properties of nanocellulose can be classified as mechanical, optical, thermal, and barrier properties.

4.2.1. Mechanical properties

The mechanical properties of nanocellulose are comparatively better than their lignocellulosic materials due to their uniform morphology. It has an average modulus of 100 GPa which is far higher than its original lignocellulosic biomass source (Dufresne 2013). The mechanical properties of nanocellulose depend on its structure i.e. crystalline or amorphous. The crystalline structure contributes to its high stiffness and special elasticity whereas the amorphous one contributes to flexibility and plasticity. Hence, the stiffness and modulus of CNCs are more due to their higher crystalline regions as compared to CNF and BC fibrils. The modulus of CNCs is in the range of 100 to 200 GPa, which is very close to the values of Kevlar (60 to 125 GPa) and even theoretically stronger than steel (200 to 220 GPa) (Lin and Dufresne 2014). The mechanical properties are the most outstanding assets of nanocellulose that could be achieved by blending it in the form of fillers in polymeric nanocomposites, which enhance its durability, resilience, rigidity, barrier properties, and flame retardancy as compared to a pure polymeric material (Mishra *et al*. 2018). As compared to other nanofillers such as carbon nanotubes, the inclusion of a smaller proportion of cellulosic nanomaterial is enough for enhancements due to its huge surface area and low cost, and it makes it more appealing for various applications (Jonoobi *et al*. 2015).

4.2.2. Optical properties

Nanocellulose has particular optical properties due to its anisotropic rod-like morphology. UV–Vis spectrometer can be used to investigate the optical features of nanocellulose films through the determination of the pattern of regular light transmittance. Measurements are carried out within the wavelength range of 200–1000 nm, and regular light transmittance is generally reported at a wavelength of 600 nm (Dorez *et al*. 2014). Nanocellulose could be optically transparent when it is packed in such a way that the interstices between the nanocellulose fibers

are small enough to prevent light scattering (Nogi *et al*. 2009). However, mechanical compression results in its structural deformation, and ultimately, the material becomes opaque. Nanocellulose films formed by slow filtration, drying process, and compression are densely packed and are not optically transparent but translucent. In suspension, it behaves like a lyotropic liquid with crystalline behavior i.e. a phase transition from an isotropic liquid to crystal liquid results due to the change in concentration. This unique characteristic makes it a potential candidate to explore new applications of nanocellulose in different fields (Revol *et al*. 1994).

4.2.3. Thermal properties

The thermal properties of nanocellulose mainly depend on its extraction method and the source of lignocellulosic biomass. The nanocellulose extracted from natural cellulosic materials has excellent thermal behavior because of its crystalline nature, versatile structure, and elimination of thermally unstable lignin materials as compared to that present in original lignocellulosic biomass (Abraham *et al*. 2011; Deepa *et al*. 2015). The thermal degradation of lignocellulosic biomass involves the decomposition of hemicelluloses, pyrolysis of lignin, depolymerization, combustion, and char oxidation. In comparison to hemicellulose, pectin, and lignin, cellulose nanofibrils have better thermal performance due to high degradation onset temperature i.e. 350°C (Chen and Kuo 2011).

4.2.4. Barrier properties

The nanocellulose films have higher tortuosity due to their small size with a large surface-to-volume ratio as compared to cellulose microparticles (Lavoine *et al*. 2012). Most of the polymeric substances utilized for food wrapping are made of non-degradable materials, which become a severe environmental issue. These types of packaging materials are being abundantly used due to their low cost, easiness of processing, and excellent barrier nature. However, the usage of bio-based materials for food wrapping purposes and other different applications has become desirable in our daily lives to sustain and develop an eco-friendly environment. Cellulose is hydrophilic and absorbs water molecules when conditioned in a moist atmosphere or liquid water. However, when the cellulosic fibers are disintegrated into nanoscale dimensions, the water vapor permeability becomes low (Minelli *et al*. 2010). Whatever the method employed, or the experimental processes utilized to prepare nanocellulose, it serves as an excellent moisture barrier for food packaging biomaterials. Nanocellulose-based nanocomposite films improve the quality of food as these serve as carriers for active substances such as antimicrobials and antioxidants and hence shelf life of food is prolonged (Andresen *et al*. 2007).

5. Production of nanocellulose from lignocellulosic biomass

The global focus is currently directed toward the conversion of lignocellulosic biomass into useful products such as nanocellulose and its further processing for various applications (Siró and Plackett 2010). The nanocellulose production from lignocellulosic sources has been a hot topic of research in recent years due to its excellent mechanical properties and great potential for future applications (Velázquez *et al*. 2022). The production of cellulose nanomaterial from lignocellulosic biomass is comprised of mainly two different steps as demonstrated in Figure 1 (Phanthong *et al*. 2018). The first step involves pre-treatment which is necessary to ensure the successful isolation of the cellulosic portion (cellulose, hemicellulose, and lignin) from tightly bound polymeric constituents such as hemicellulose, lignin, and other chemicals compounds in lignocellulosic biomass. The primary purpose of this fractionation is to increase the accessibility of cellulosic material to chemical attack followed by cellulose hydrolysis to produce nanocellulose (Khan *et al*. 2023; Cabiac *et al*. 2011). During the second step, nanocellulose is obtained from cellulosic fibrils using various methods of extraction (Phanthong *et al*. 2018; Haafiz *et al*. 2014). A detailed discussion of both steps is given below in the following sections:

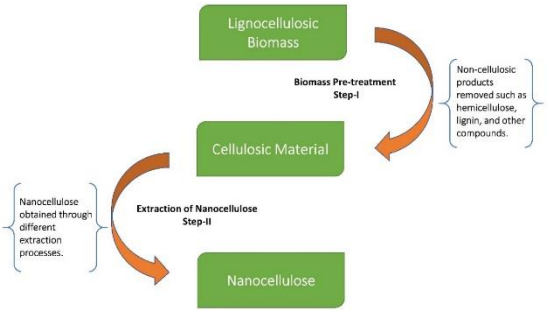


Figure 1. Production of nanocellulose from lignocellulosic biomass

5.1. Biomass pre-treatment

Nanocellulose is produced from lignocellulosic materials that are abundantly present on Earth. As discussed earlier, lignocellulosic biomass is composed of cellulose and non-cellulosic substances such as hemicellulose, lignin, and other compounds (extractives and inorganic substances). Biomass pre-treatment is a significant step in eliminating non-cellulosic products so that the remaining cellulosic materials can be used for further nanocellulose extraction (Haafiz *et al*. 2014; Ibarra *et al*. 2021). The two classical approaches used for biomass pre-treatment include the acid-chlorite and alkaline treatment processes. The acid-chlorite treatment process, also known as the bleaching or delignification process has been extensively applied in pulp industries (Cullen and MacFarlane 2005; Hubbell and Ragauskas 2010). In this process, lignocellulosic biomass is mixed with distilled water, sodium chlorite, and acetic acid and stirred at 70–80°C for 4–12 h (Cullen and MacFarlane 2005; Mandal and Chakrabarty 2011; Johar *et al*. 2012; dos Santos *et al*. 2013). At different intervals, pH is controlled by adding acetic acid and sodium chlorite to the mixture. Distilled water is added to the stirring mixture until the pH becomes neutral. The obtained solid product as fibers mainly consists of cellulose and hemicellulose and is known as holocellulose, which is then

dried in the oven at 50°C. The white color of holocellulose fiber suggests that the lignin and other impurities have successfully been removed (Johar *et al*. 2012; dos Santos *et al*. 2013; Phanthong *et al*. 2015).

The alkaline treatment is used to eliminate the amorphous polymers and the rest of the lignin (Mandal and Chakrabarty 2011; Johar *et al*. 2012; dos Santos *et al*. 2013). During this treatment, sodium hydroxide (4–20% w/v) is mixed with holocellulose for 1–5 h, and the obtained solid products are washed with distilled water until the pH becomes neutral (Cullen and MacFarlane 2005; Hubbell and Ragauskas 2010; Mandal and Chakrabarty 2011; Johar *et al*. 2012). It is then dried in an oven at 50°C, and the obtained solid product mainly consists of cellulose (Phanthong *et al*. 2015). Many research groups have used these pre-treatment methods to eliminate non-cellulosic materials from lignocellulosic biomass as demonstrated in Figure 2.

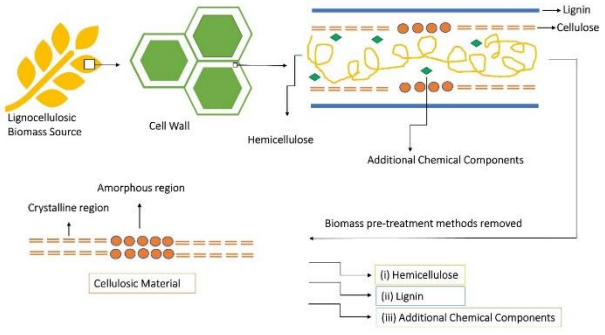


Figure 2. Schematic representation of lignocellulosic biomass pre-treatment

5.2. Production of nanocellulose

In general, many research groups have discussed that the production procedure involves three different processes i.e. acid hydrolysis, enzymatic hydrolysis, and mechanical process (Figure 3). As the cellulose chain consists of ordered and disordered regions, acid can efficiently hydrolyze the disordered parts and the ordered regions left as the nanocellulose (Mehmood *et al*. 2023; Lavoine *et al*. 2012). The properties of extracted nanocellulose depend on the temperature, acid concentration, and reaction time during acid hydrolysis (Khan *et al*. 2023; Lavoine *et al*. 2012; Naveed *et al*. 2020). Sodium hydroxide is used to neutralize the pH of the obtained products (Wang *et al*. 2007).

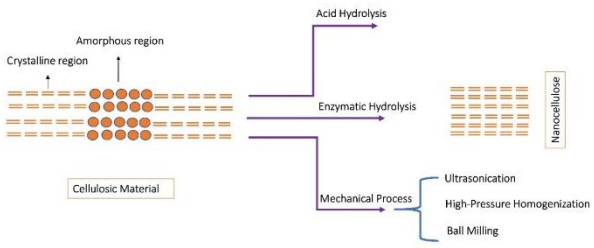


Figure 3. Schematic representation of production of nanocellulose

In enzymatic hydrolysis, enzymes are used to digest or modify the cellulose fibers (Khalil *et al*. 2014; Zielińska *et al*. 2021). In general, this process can be carried out under mild conditions due to the presence of enzymes; however, a long time of exposure is required (Khan *et al*. 2023; Lavoine *et al*. 2012). This problem could be solved by incorporating enzymatic hydrolysis with other methods.

During the mechanical process, high shear force in the longitudinal axis is used to break cellulosic fibers into cellulose nanofibrils (Khalil *et al*. 2012; Dufresne 2013; Khalil *et al*. 2014). The mechanical process includes ultrasonication, high-pressure homogenization (HPH), and ball milling. During ultrasonication, the hydrodynamic force of high-intensity ultrasonication is used to defibrillate cellulosic fiber (Dufresne 2013; Khalil *et al*. 2014; Zielińska *et al*. 2021; Szymańska-Chargot *et al*. 2022). When the liquid molecules are subjected to absorb ultrasonic energy, the mechanical oscillating power is generated, which results in the formation, expansion, and collision of microscopic gas bubbles (Filson and Dawson-Andoh 2009; Zhou *et al*. 2012). The high-pressure homogenization is carried out by passing cellulose slurry through a vessel under high pressure and velocity (Khalil *et al*. 2014).

During ball milling, the centrifugal force from the rotating jar with shear forces among balls and between balls and the surface of the jar is applied to defibrillate cellulose fibers (Baheti *et al*. 2012; Feng *et al*. 2004). The cellulose fibrils are broken down into smaller cellulosic materials (Avolio *et al*. 2012; Kim *et al*. 2013; Barakat *et al*. 2014). However, the main drawback of this method is high energy consumption, which could be decreased by combining it with other pretreatment methods (Khalil *et al*. 2014). The limitation of conventional, and green mechanical techniques and the advantages of emerging technologies for nanocellulose extraction from lignocellulosic biomass is presented in Figure 4.

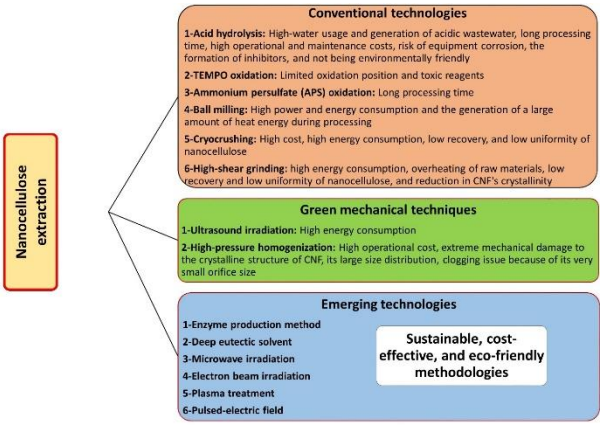


Figure 4. Conventional, green mechanical, and emerging technologies for nanocellulose extraction/production from lignocellulosic biomass

6. Applications of nanocellulose

Nanocellulose has been an attractive material with multiple applications due to its extraordinary properties (Figure 5). A detailed description of each application is given below:

6.1. Composite industry

In recent years, the use of nanocellulose in the field of polymer reinforcement to create high-performance biomaterials has become the principal subject of interest. It is due to its nano-size, high aspect ratio, good dispersion in hydrophilic systems, higher uniformity with fewer defects, and enhanced mechanical properties (Spence *et al*. 2011). It could be applied as a reinforcement material to synthesize nanocomposites with water-soluble polymer solutions to improve the mechanical strength and viscosity of dry composites.

One of the most interesting uses is its addition to biodegradable polymers, which not only enhances the mechanical properties of the polymers but also enhances their biodegradation (García-González *et al*. 2011; Jung *et al*. 2015). It has been a model nanofiller in many polymeric nanocomposite systems as it can improve mechanical strength and modulus even at very low concentrations. Nanocellulose-based polymeric composites can be utilized to develop membranes, textiles, fibers, electroactive polymers, batteries, supercapacitors, sensors, and actuators that utilize electromechanical responses (Hoeger *et al*. 2011; Yang *et al*. 2012). Its incorporation into different materials can improve mechanical strength, optical and thermal stability, and barrier properties because of its better interfacial interaction and crystallinity.

Nanocellulose-based biodegradable nanocomposite films can also be used in food wrapping where lower permeability to moisture, aroma, gases, and oil is required (Lange and Wyser 2003).

6.2. Biomedical industry

Nanocellulose is also used in the field of medical sciences due to its good biocompatibility, low toxicity, renewability, and outstanding physical features. Pure nanocellulose is harmless to humans and, therefore, can be used in healthcare products such as personal hygiene products, biomedicines, and cosmetics. It can also be used as an excellent carrier for drugs and the immobilization of enzymes after chemical modification (Liu *et al*. 2015; Lin and Dufresne 2014; Jorfi and Foster 2015). Other interesting medical applications of nanocellulose include soft tissue implants, blood vessel replacements, and many other useful applications that are still under investigation in recent years (Lin and Dufresne 2014; Jorfi and Foster 2015). Nanocellulose-based aerogels are also gaining attention in the field of biomedicine and pharmaceutical industries due to the unique structure and higher surface area of nanocellulose, which improves drug bioavailability and drug-loading capacity (García-González *et al*. 2011). Cellulose nanomaterials have also been used in wound healing patches, biosensors, bioprobes, tissue engineering scaffolds, fluorescence bioassays, bioimaging, and hydrogels for medical and pharmacological applications (Dong and Roman 2007)

6.3. Paper and paperboard industry

Nanocellulose also plays an essential role in the paper and paperboard manufacturing sector due to its significant reinforcing effect on paper materials (Ioelovich and Leykin 2004). It enhances the fiber-fiber bond strength and exhibits admirable mechanical strength even 2–5 times more than common papers produced from traditional refining processes (Henriksson *et al*. 2008). It has been observed that the application of nanocellulose as a coating agent on paper and paperboard surfaces enhances the barrier properties, in particular, the air resistance (Kumar *et al*. 2016). It improves the structural properties (flexibility and transparency) of paperboard to be used for electronic devices (Jung *et al*. 2015). Modified nanocellulose-based paper is transparent, optically clear, and foldable, and can be used in areas that require surfaces with anti-corrosion and self-cleaning ability. Such types of transparent papers can also be applied instead of traditional papers in electronic devices such as solar cells, flexible circuits, and flexible displays.

6.4. Other potential applications

Nanocellulose can also be used as a texturing agent in food, CO_2 adsorbent, thickener in cosmetics, filler in some textiles, oil recovery, and many other fields (George and Sabapathi 2015; Gebald *et al*. 2011; Kaushik and Moores 2016; Wei *et al*. 2016). It has also been used in the field of emulsion and dispersion owing to its ability to form stable colloidal dispersions in the presence of water and is considered suitable for water-soluble polymers such as latexes.

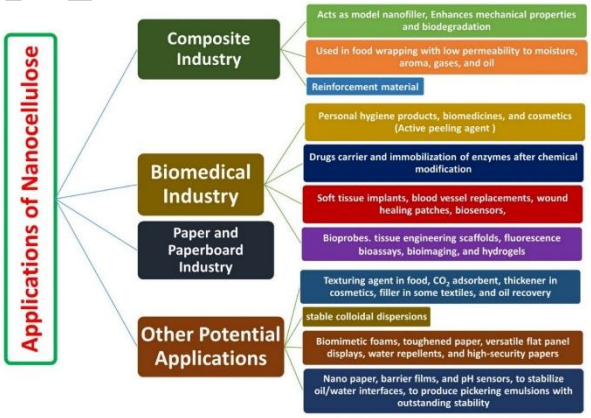


Figure 5. Applications of nanocellulose in various fields

Though stable dispersions in the presence of nonpolar solvents can also be achieved by using either surfactants or surface chemical grafting, however, it is also possible to disperse nanocellulose in nonpolar solvents without surfactant or chemical modification (Samir *et al*. 2005; de Jesus Carvalho de Souza *et al*. 2021; Xu *et al*. 2021; Sun *et al*. 2022). Besides good dispersion, nanocellulose, in some cases, forms a percolated network within a polymer matrix, which improves the features of polymeric nanocomposites. Such kinds of nanocomposites are used to produce biomimetic foams, toughened paper, versatile flat panel displays, water repellents, and high-security papers (Nogi *et al*. 2009; Henriksson *et al*. 2008; Svagan *et al*. 2008; Bayer *et al*. 2011). Other distinctive applications of nanocellulose are its use in a range of products such as nano paper, barrier films, and pH sensors, to stabilize oil/water interfaces, and to produce Pickering emulsions with outstanding stability (Henriksson *et al*. 2008; Nielsen

et al. 2010; Belbekhouche *et al*. 2011; Kalashnikova *et al*. 2012).

7. Nanocellulose in waterborne wood coatings

The coatings not only play a significant role in the beautification and glossiness of material but also protect from the deleterious effects of microorganisms and acid rain (Nikolic *et al*. 2015). However, traditional coatings are mostly solvent- or oil-based and are not environment hence releasing volatile organic compounds (VOCs) into the environment. Thus, waterborne coatings, which release hardly any VOCs, are a suitable replacement for traditional organic solvent-based coatings. These coatings are advantageous due to their lower VOC emissions, higher peel strength, good low-temperature ductility, and low viscosity (Mazela *et al*. 2022). However, due to the low mechanical performance (hardness, scratch resistance) of waterborne coatings, their application in the waterborne coating industry has become limited (Xu *et al*. 2013; Vardanyan *et al*. 2014; Veigel *et al*. 2014; Noreen *et al*. 2016).

The impact of value addition with different forms of nanocellulose on the wood coatings' properties and their mechanisms have been summarized in Table 1. To enhance the mechanical performance of waterborne wood coatings, nanofillers such as nano-titanium dioxide, nano-zinc oxide, nano-silica, and clay have been widely used due to their small size and surface effect.[169-173] Moreover, nanocellulose as fillers in the form of CNFs and CNCs could be an excellent reinforcement filler in waterborne wood coatings due to its high modulus and potential for sustainable production (Khan *et al*. 2022; Veigel *et al*. 2014; Grüneberger *et al*. 2014; Poaty *et al*. 2014; Kluge *et al*. 2017; Yang *et al*. 2020).

Nanocellulose has received tremendous attention in the sustainable biomaterial-based industry due to its unique nature. Coating technology is mostly applied in the construction, furniture, automobiles, military, and packaging industries (Zhou *et al*. 2021). Nanocellulose improves service life and plays an essential role in its functionality. Regarding its use in coatings, several studies have been published, but its use in waterborne wood coatings is still in progress, and limited research work has been reported. Nanocellulose has improved resistance against scratching and abrasion of acrylic latex, high-solids coating, and UV-curing waterborne varnishes (Vardanyan *et al*. 2014; Kaboorani *et al*. 2016; Poaty *et al*. 2014; Landry *et al*. 2008). Besides developments in the field of coating, also optical properties may be affected by the addition of nanocellulose to coatings (Vlad-Cristea *et al*. 2013). It is reported that it can be straightforward to disperse in waterborne coatings; however, chemical hydrophilization would be required for nonpolar systems such as an organofunctional silane or acryloyl chloride, or alkyl quaternary ammonium bromides (Poaty *et al*. 2014; Landry *et al*. 2008).

8. Conclusions and future perspectives

The main focus of this review is to overview the process of production of nanocellulose (CNCs, CNFs, and BC) from lignocellulosic biomass through different processes and discuss its potential applications. Several research projects have worked on the production of nanocellulose from various sources of lignocellulosic biomass and it is still the main challenge to make it easily available on a commercial scale to meet its demand due to its significant features. It is a material with carbon-neutral, sustainable nature, eco-friendly, recyclable, and harmless nature. Hence, it has special morphology and geometric proportions, high specific surface area, crystallinity, liquid crystalline behavior, rheological properties, mechanical stability, alignment and orientation, barrier properties, chemical reactivity of the surface, lack of toxicity, biocompatibility, biodegradability, many more others special features which make it a potential candidate to generate new applications of nanocellulose in the field of science and technology. It has played a vital role in various industries such as composite, biomedical, paper, and pulp industries, and many others. It also has the potential to replace synthetic or petrochemical-based materials to enhance the environmental footprint of many of these industries. While the subject of nanocellulose has been researched intensively over the last few years, still the place for new advancements exists such as its production from novel sources, and its application in various fields should attract more attention in future studies. Due to its cost-effectiveness and environmentally friendly nature, further expansion in the limits of nanocellulose applications, especially in the field of material science and engineering, would be of particular interest in the future for scientists and technologists around the globe. Similarly, the production of nanocellulose is still expensive and requires energy-intensive procedures, and could be resolved by upscaling the manufacturing and economical surface modification methods. Overcoming all these obstacles may further expand nanocellulose's role in enhancing material science and engineering while also promoting sustainability.

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Table 1. Impact of value addition with different forms of nanocellulose on the wood coatings properties and their mechanisms

Coating	Value addition	Effect on coating properties	Mechanisms	References
Polyvinyl alcohol (PVA) coating	Heating treatment and lignin nanocellulose	The composite coating had improved corrosion resistance. The elimination reaction due to heating significantly reduced the water transport channels.	Elimination reaction, the lignin nanocellulose worked effectively as a barrier against the penetration of corrosive media	(Zhang *et al.* 2021)
Salvia-based waterborne polyurethane-urea (WBPUU)	Cellulose nanocrystals (CNC)	CNC addition modulates the soft and hard phase segregation	Effective CNC incorporation without agglomeration	(Santamaria-Echart *et al.* 2021)
Waterborne polyurethane (WPU) coatings	Nanocellulose crystalline (NCC) and silver nanoparticles	Improved adhesion of the NCC-WPU composites. The antibacterial effect was at its best with the addition of NCC (0.5%) and the proportion of silver elements (5%)	Improved adhesion and antibacterial effect	(Cheng *et al.* 2020)
Waterborne blocked polyisocyanate (PIC).	CNC	Exhibited a reversible humidity-dependent color change which can be used in humidity sensor applications. At 90% relative humidity, the hygroscopic strain was 20 times less compared to untreated CNCs	Reduction in hygroscopic strain	(Chowdhury *et al.* 2020)
PVA coating	Polyethylene glycol methacrylate (MPEG) and cellulose nanocrystals	Strong reinforcing effect with high transparency	Colloidal stability	(Mabrouk *et al.* 2020)
Modified vegetable oil-based WPU	Reacting excess amino group on the POSS with the -NCO group	Increased initial decomposition temperature, water contact angle, and tensile strength	Improvement in mechanical properties, thermal stability, and water resistance	(Zhang *et al.* 2020)
Waterborne acrylate-based matrix coating	Cellulose nanocrystals	Improved barrier performance	Barrier performance	(He *et al.* 2019)
Waterborne acrylate formulation	CNC or cellulose nanofibrils (CNF)	CNC-based barrier coating produced had high stable polarization resistance and open circuit potential	Stable polarization resistance, open circuit potential	(He *et al.* 2020)
WPU paint	Nanocellulose	Improved hardness and abrasion resistance, tensile strength, and elongation at break	Improvement in comprehensive properties	(Kong *et al.* 2019)
Photopolymerizable siloxanemethacrylic-based resin/CNC	Nanocellulose and microcellulose	Improved hydrophobicity, thermal properties, stiffness into the neat cured systems with no embrittling, surface hardness with reduced water sorption	Improvement in comprehensive properties	(Cataldi *et al.* 2017)
Epoxy resin-based nanocomposites	Nanocellulose loadings	Improved anti-corrosion properties	Improved physicochemical and anti-corrosion properties	(Ma *et al.* 2017)
Composite of TEMPO-oxidized cellulose nanofibers (TOCNs) and WPU-based coating	TOCNs	Enhanced the mechanical properties	Improvement in mechanical properties	(Cheng *et al.* 2016)
CNC modified by a cationic	CNC was modified by a	CNC improved the barrier and optical properties and	Improvement in the barrier and	(Kaboorani *et*

surfactant was added to the coating system	cationic surfactant at two loadings (1 and 3%).	agglomeration	optical properties	*al.* 2016)
Composite of CNFs with γ-aminopropyltriethoxysilane (APS)	γ-aminopropyltriethoxysilane	Demonstrated distributing uniformly, retained high light transmittance, and improved mechanical properties.	High light transmittance with improved mechanical properties	(Tan *et al.* 2016)
Dispersion and stabilization impact of zinc oxide (ZnO) nanoparticles in waterborne wood coatings.	Nanofibrillated cellulose (NFC) and zinc oxide (ZnO)	Improved unstabilized ZnO distribution and prevented sedimentation of ZnO, improved film formation, and inhibited crack formation. NFC had a pronounced matting effect	Improved distribution of unstabilized ZnO in the coatings, film formation, and inhibition of crack formation	(Grüneberger *et al.* 2015)
Composite films of NFC, and commercial acrylic and alkyd polymeric binders	Acrylic and alkyd polymeric binders	Improved reinforcing effect	Improved mechanical properties	(Grüneberger *et al.* 2014)
Carbon chains grafting of cellulose nanocrystal (CNC)	Alkyl quaternary ammonium bromides or acryloyl chloride	CNC derivatives were better dispersed, conferring a higher scratch resistance	CNC dispersion and mechanical properties	(Poaty *et al.* 2014)
CNC was mixed in thevarnishes	Varnishes	Improved wear resistance (30-40%)	Mechanical properties	(Vardanyan *et al.* 2014)
Microfibrillated cellulose (MFC) and nanocrystalline cellulose (NCC) were added to waterborne	MFC and NCC	Lowered gloss, improved hardness, and modulus	Improvement in mechanical properties	(Veigel *et al.* 2014)
Commercially available paperboards were coated with thin layers of nanocellulose	Commercially available paperboards	Improved water vapor barrier properties	Water vapor barrier properties	(Aulin and Ström 2013)
ZnO and polymer composites	CNC	Reduced destructive effects of UV light and humidity	Reduced ΔE value	(Harandi *et al.* 2023)

Author contributions

"Conceptualization, M.N.K., A.D., A.S., Z.H.F., N.R., E.A., M.I.D., M.T., R.I., S.M.E., I.A., and S.B.; software, M.N.K., A.D., A.S., Z.H.F., N.R., E.A., M.I.D., M.T., R.I., S.M.E., I.A., and S.B.; validation, M.N.K., A.D., A.S., Z.H.F., N.R., E.A., M.I.D., M.T., R.I., S.M.E., I.A., and S.B.; writing—original draft preparation, M.N.K., and A.D.; writing—review and editing, M.N.K., A.D., A.S., Z.H.F., N.R., E.A., M.I.D., M.T., R.I., S.M.E., I.A., and S.B.; visualization, M.N.K., A.D., A.S., Z.H.F., N.R., E.A., M.I.D., M.T., R.I., S.M.E., I.A., and S.B.; supervision, A.S., Z.H.F., N.R., and M.I.D.; project administration, A.S., Z.H.F., N.R., and M.I.D.; funding acquisition, A.D., R.I., S.M.E., I.A., and S.B.

Conflicts of Interest

"The authors declare no conflict of interest."

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