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

review

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18 GRAPHICAL ABSTRACT

Conventional technologies

1-Acid hydrolysis: High-water usage and generation of acidic wastewater, long processing time, high operational and maintenance costs, risk of equipment corrosion, the formation of inhibitors, and not being environmentally friendly

2-TEMPO oxidation: Limited oxidation position and toxic reagents

3-Ammonium persulfate (APS) oxidation: Long processing time

4-Ball milling: High power and energy consumption and the generation of a large amount of heat energy during processing

5-Cryocrushing: High cost, high energy consumption, low recovery, and low uniformity of nanocellulose

6-High-shear grinding: high energy consumption, overheating of raw materials, low recovery and low uniformity of nanocellulose, and reduction in CNF's crystallinity

Green mechanical techniques

1-Ultrasound irradiation: High energy consumption

2-High-pressure homogenization: High operational cost, extreme mechanical damage to the crystalline structure of CNF, its large size distribution, clogging issue because of its very small orifice size

Emerging technologies

- 1-Enzyme production method
- 2-Deep eutectic solvent
- 3-Microwave irradiation
- 4-Electron beam irradiation
- 5-Plasma treatment
- 6-Pulsed-electric field

Sustainable, costeffective, and eco-friendly methodologies

Nanocellulose extraction

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22 Abstract

Lignocellulosic biomass is a complex natural polymer primarily composed of cellulose, 23 hemicellulose, lignin, and various other chemical components. The cellulose in the lignocellulosic 24 biomass can depolymerize into a nano-dimension biomaterial called nanocellulose, which 25 possesses unique features with potential application in various fields. Nanocellulose production 26 27 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 28 scientists and technologists worldwide. This production faces many challenges in utilizing the 29 cellulose from lignocellulosic biomasses and subsequent processing for their conversion into 30 nanocellulosic materials and their further applications in various fields of science and technology. 31 This current review not only focuses on the production of nanocellulose from lignocellulosic 32 biomass through different production methods but also discusses various sources, types, 33 properties, and their applications in material science and biomedical engineering. This research 34 review certainly shows that in the future, nanocellulose has great potential to be used as a 35 renewable source in the field of sustainable materials and nanocomposites. 36

37 Keywords: lignocellulosic biomass; cellulose; nanocellulose; production and applications

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39 **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.^{1,2} Compared to synthetic materials, bio-based materials are easily decomposable, regenerated naturally, and have no adverse effects on the environment.^{3,4} These biopolymers are abundant in nature with numerous applications potential in material sciences.⁵⁻⁹ 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.¹⁰

The cellulose from a natural source contains both crystalline (proper arrangement of molecules) 48 as well as amorphous (lack of arrangement of molecules) structures. The extent of crystallinity and 49 amorphous structure mainly depends on the source from which the corresponding cellulose is 50 extracted, purity, time, and temperature.¹¹ When cellulose molecules are subjected to a set of 51 experimental conditions like mechanical, chemical, and enzyme actions, it leads to the production 52 of crystalline structures called cellulose nanocrystals (CNCs).¹² The bulk form of cellulose has 53 higher amorphous fractions than CNCs. The cellulose nanocrystals are composed of rod-shaped 54 molecules in a proper arrangement, which builds up the crystalline structure. The synonym 55 terminologies used for CNCs are whiskers, nanocellulose, nanofibers, and microcrystalline, but 56 CNCs are the most appropriate and widely accepted nomenclature used.¹³ The crystalline nature 57 of CNCs is responsible for a specific strength, biodegradability, useful rheological behavior, 58 modulus, high surface area, maximum chances of chemical modification, biocompatibility, 59 lightweight, high aspect ratio, outstanding oxygen permeation resistance, oil grease barrier, water 60 vapor transmission rate resistance, aqueous liquid barrier, and unique liquid crystalline properties. 61 ¹⁴⁻¹⁶ Many researchers have produced CNCs from various cellulosic sources e.g. durian rind, wheat 62 bran, corn stover, sugarcane bagasse, sago seed shells, raw cotton linter, sisal fibers, mengkuang 63 leaves, kusha grass, mulberry, cotton stalks, and many others.¹⁷⁻²⁸ In comparison to traditional 64 composite materials, CNCs exhibit high mechanical, electrical, optical, thermal, and gas barrier 65 properties.^{26,29-32} Therefore, the global scientific community is focusing on the use of CNCs for 66 67 various industrial applications like packaging, automobile, thermal management, and optical

68 industries.^{28,33-35} Due to their exceptional properties, CNCs have been used as a reinforcement 69 component in nanocomposites^{36,37}, barriers in packaging applications³⁸, transparent media in 70 organic electronics, and anti-counterfeiting in security applications.³⁹ The CNCs can also be 71 utilized in film, aerogel, and foam, depending on the required application.²⁸

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.^{15,40-42} 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.^{15,43-46}

Another distinctive application of CNCs is their use in coatings.⁴⁷ Due to their high stiffness, 79 strength, and barrier properties, these offer enhanced mechanical, scratch-resistant, gas/water 80 barrier, and weathering-resistant properties to the coatings. The traditional coatings are mostly oil-81 or solvent-based, which release volatile organic compounds (VOCs), thereby polluting the 82 environment. The use of CNCs in different coatings such as waterborne coatings is 83 environmentally friendly and therefore, has become the focus of intense research for scientists and 84 technologists worldwide. Based on the above discussion, CNCs have great potential to be used in 85 the field of coatings due to their state-of-the-art properties. The present review summarizes the 86 87 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 88 89 directions by finding research gaps.

90 2. Lignocellulosic biomass

Lignocellulosic biomass is an abundantly available complex biopolymer composed of cellulose, 91 hemicellulose, and lignin with a small number of inorganic materials and extractives.^{48,49} These 92 polymeric constituents are related to each other in a heater matrix to different degrees and varying 93 compositions, depending on the species and even the source of the biomass from a species.⁵⁰ The 94 worldwide annual production of lignocellulosic biomass is approximately 1.815×10^{11} metric 95 tons.⁵¹ Due to its higher availability and renewable nature, it has been one of the world's most 96 potential resources having diverse applications. However, various pre-treatment steps are required 97 for the successful separation of the cellulosic portion from tightly bound polymeric constituents in 98 lignocellulosic biomass. The presence of cellulose in lignocellulosic biomass is capable of 99 depolymerization into a biomaterial with nano-dimensions, excellent mechanical properties, rich 100 in hydroxyl groups available for modification, high surface area, and natural features with eco-101 friendly applications in the biomedical field, reinforcement of nanocomposite material, 102 pharmaceutical carriers, electronic substrates, and many others.⁵² 103

104 2.1. Cellulose

Cellulose is one of the natural, renewable, and biodegradable polymers that is abundantly available 105 on Earth.^{7,53,54} It is an essential structural component of the cell wall present in many plants, 106 107 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 108 that ~ 1.5×10^{12} tons of cellulose are synthesized each year around the globe.⁵⁵ Cellulose is a 109 natural linear polymer (polysaccharide), composed of β -D-glucopyranose units connected via β -110 1-4-linkages.⁵⁶ The cellulose present in nature does not exist in the form of an isolated individual 111 molecule, but it occurs as assemblies of individual cellulose chain-forming fibers. These fibrils are 112 113 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.⁵⁷

Common sources of cellulose include cotton linters, jute, and wood pulp. Cotton linters contain 117 approximately 90-95% cellulose, while in jute and wood pulp, it is around 45-63 and 40-45%, 118 respectively.⁵⁸ Cellulose extracted from highly pure cotton linters requires minimal treatment 119 (NaOH treatment) to remove non-cellulosic materials. However, other fibers such as jute, wood 120 pulp, hemp, and ramie need extensive processing to remove the non-cellulosic components. 121 Cellulose extraction from lignocellulosic biomass is a topic of intense research with the advent 122 and advancement of nanotechnology. It attracts further interest in the new form of "nanocellulose" 123 to be used as a novel and advanced material and has become the principal theme of research work 124 carried out by scientists and technologists worldwide.59 125

126 2.2. Hemicellulose

Hemicelluloses are heterogeneous polysaccharides present in the cell walls of almost all plants and 127 act as a linkage molecule between cellulose and lignin. Hemicellulose polysaccharide chains are 128 relatively short with an average degree of polymerization of about 200. Hence, these can easily be 129 hydrolyzed and extracted from plant tissue by using water, acid, or aqueous alkali. Unlike 130 cellulose, it is an amorphous polymer with little strength.^{59,60} Hemicelluloses like many other 131 polysaccharides, act as substantial barriers to oxygen. However, these are moisture-reactive and 132 133 show low barrier resistance under humid conditions. Mannan-type hemicelluloses have successfully been used in the preparation of packaging films, showing strong film-forming 134 properties and resistance to oxygen diffusion.⁶¹ 135

136 *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 phydroxyphenyl, guaiacyl, and syringyl units.⁶² 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.^{63,64}

143 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.⁶⁵ 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.³³

150 **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.^{15,66} A brief description of the significant cellulose sources is given below:

154 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.^{67,68} A large quantity of cellulose can also be obtained from other plants such as grasses, aquatic plants, ramie, jute, sisal, hemp, flax, etc.^{8,69} Agricultural wastes such as rice and wheat straw, sawdust, cotton stubbles, sugarcane bagasse, etc. are also among the
significant sources of cellulose.^{15,70,71}

161 *3.2. Algae*

162 Cellulose is the fundamental component of the cell wall of many algal species. Among algal 163 species, blue-green algae is a major source of cellulose while red and yellow algal species contain 164 smaller amounts of cellulose. The cellulose extracted from valonia and cladophora species is 165 crystalline (95%). The properties of microfibrils depend on the nature of the cellulose biosynthesis 166 process occurring among different algal species.⁷²

167 *3.3. Bacteria*

168 Komagataeibacter xylinus is a well-known bacterial species that produces cellulose using nitrogen 169 and carbon to create dense and clear cellulose microfibrils.⁷³ The cellulose extracted from 170 microbes is unique possessing outstanding properties due to its nanostructure, purity, and good 171 mechanical strength compared to the cellulose derived from plants.⁵⁵

172 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.^{13,59,74}

177 **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.^{75,76} The stiffness of nanocellulose is very high, and it is up 182 to 220 GPa of elastic modulus, which is higher than Kevlar fiber. It has an excellent tensile strength 183 value of up to 10 GPa which is superior to the value of cast iron. It also has a much higher ratio of 184 strength to weight than stainless steel. It is rich in very reactive hydroxyl groups on its surface that 185 could be modified to give different surface properties.^{8,77,78} Because of its biodegradable and 186 renewable nature, mechanical strength, crystallinity, high specific surface area, tailorable surface 187 chemistry, anisotropic shape, biocompatibility, and optical properties, nanocellulose has 188 extensively been used in various scientific disciplines such as material sciences and biomedical 189 engineering.^{79,80} Moreover, nanocellulose has some other important features, which have been 190 extensively applied in the fields of packaging, filtration, electronic sensors, hydrogel, 191 reinforcement of nanocomposites, paper and pulp, paints, coatings, and many others.81 192

193 *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.⁸²

198 *4.1.1. Cellulose Nanofibrils (CNFs)*

199 Cellulose nanofibrils (CNFs) are cellulosic nanomaterials, which consist of linear fibrils having a 200 diameter ranging from 5 to 50 nm and a length of a few micrometers.^{83,84} CNFs comprise both the 201 amorphous and crystalline regions in a single fiber⁸⁵ and can be extracted from cellulosic fibers by 202 using three different kinds of processes such as mechanical treatments, chemical treatments, and a 203 combination of mechanical and chemical treatments.⁷⁷ In mechanical treatment, CNFs are synthesized through high-pressure grinding of cellulosic pulp suspension, hence strong entangled
 networks of nanofibrils are formed.⁸⁶

206 *4.1.2. Cellulose Nanocrystals (CNCs)*

207 Cellulose nanocrystals (CNCs) are cellulose nanomaterial with a length of 200–500 nm and a 208 diameter of 3–35 nm. These are produced through acid hydrolysis of cellulosic substances.⁸⁷ In 209 this method, concentrated sulfuric acid is used to dissolve the amorphous cellulosic material into 210 crystalline.⁸⁸ Hence, rod-like CNCs with nearly perfect crystallinity and almost 90% purity are 211 obtained.

212 *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.⁸⁹ 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.⁹⁰

218 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.^{15,91,92} The properties of nanocellulose can be classified as mechanical, optical, thermal, and barrier properties.

223 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.⁷⁸ The mechanical properties of

nanocellulose depend on its structure i.e. crystalline or amorphous. The crystalline structure 227 contributes to its high stiffness and special elasticity whereas the amorphous one contributes to 228 flexibility and plasticity. Hence, the stiffness and modulus of CNCs are more due to their higher 229 crystalline regions as compared to CNF and BC fibrils. The modulus of CNCs is in the range of 230 100 to 200 GPa, which is very close to the values of Kevlar (60 to 125 GPa) and even theoretically 231 stronger than steel (200 to 220 GPa).⁸³ The mechanical properties are the most outstanding assets 232 of nanocellulose that could be achieved by blending it in the form of fillers in polymeric 233 nanocomposites, which enhance its durability, resilience, rigidity, barrier properties, and flame 234 retardancy as compared to a pure polymeric material.⁹³ As compared to other nanofillers such as 235 carbon nanotubes, the inclusion of a smaller proportion of cellulosic nanomaterial is enough for 236 enhancements due to its huge surface area and low cost, and it makes it more appealing for various 237 applications.⁷⁴ 238

4.2.2. Optical Properties 239

Nanocellulose has particular optical properties due to its anisotropic rod-like morphology. UV-240 Vis spectrometer can be used to investigate the optical features of nanocellulose films through the 241 determination of the pattern of regular light transmittance. Measurements are carried out within 242 the wavelength range of 200–1000 nm, and regular light transmittance is generally reported at a 243 wavelength of 600 nm.⁹⁴ Nanocellulose could be optically transparent when it is packed in such a 244 way that the interstices between the nanocellulose fibers are small enough to prevent light 245 scattering.95 However, mechanical compression results in its structural deformation, and 246 ultimately, the material becomes opaque. Nanocellulose films formed by slow filtration, drying 247 248 process, and compression are densely packed and are not optically transparent but translucent. In 249 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.¹¹

253 4.2.3. Thermal Properties

The thermal properties of nanocellulose mainly depend on its extraction method and the source of 254 lignocellulosic biomass. The nanocellulose extracted from natural cellulosic materials has 255 excellent thermal behavior because of its crystalline nature, versatile structure, and elimination of 256 thermally unstable lignin materials as compared to that present in original lignocellulosic 257 biomass.^{96,97} The thermal degradation of lignocellulosic biomass involves the decomposition of 258 hemicelluloses, pyrolysis of lignin, depolymerization, combustion, and char oxidation. In 259 comparison to hemicellulose, pectin, and lignin, cellulose nanofibrils have better thermal 260 performance due to high degradation onset temperature i.e. 350 °C.98 261

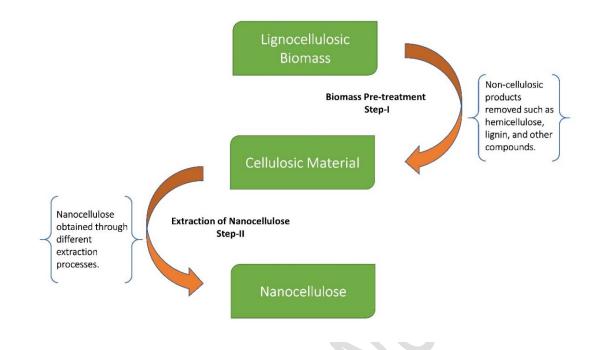
262 4.2.4. Barrier Properties

The nanocellulose films have higher tortuosity due to their small size with a large surface-to-263 volume ratio as compared to cellulose microparticles.³⁸ Most of the polymeric substances utilized 264 for food wrapping are made of non-degradable materials, which become a severe environmental 265 266 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 267 wrapping purposes and other different applications has become desirable in our daily lives to 268 269 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 270 fibers are disintegrated into nanoscale dimensions, the water vapor permeability becomes low.⁹⁹ 271 272 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.¹⁰⁰

276 5. Production of Nanocellulose from Lignocellulosic Biomass

The global focus is currently directed toward the conversion of lignocellulosic biomass into useful 277 products such as nanocellulose and its further processing for various applications.¹⁰¹ The 278 nanocellulose production from lignocellulosic sources has been a hot topic of research in recent 279 years due to its excellent mechanical properties and great potential for future applications.¹⁰² The 280 production of cellulose nanomaterial from lignocellulosic biomass is comprised of mainly two 281 different steps as demonstrated in Figure 1.80 The first step involves pre-treatment which is 282 necessary to ensure the successful isolation of the cellulosic portion (cellulose, hemicellulose, and 283 lignin) from tightly bound polymeric constituents such as hemicellulose, lignin, and other 284 chemicals compounds in lignocellulosic biomass. The primary purpose of this fractionation is to 285 increase the accessibility of cellulosic material to chemical attack followed by cellulose hydrolysis 286 to produce nanocellulose.^{28,103} During the second step, nanocellulose is obtained from cellulosic 287 fibrils using various methods of extraction.^{80,104} A detailed discussion of both steps is given below 288 289 in the following sections:



290

291 Figure 1: Production of nanocellulose from lignocellulosic biomass

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293 5.1. Biomass Pre-treatment

294 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 295 such as hemicellulose, lignin, and other compounds (extractives and inorganic substances). 296 297 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.^{104,105} The two 298 classical approaches used for biomass pre-treatment include the acid-chlorite and alkaline 299 treatment processes. The acid-chlorite treatment process, also known as the bleaching or 300 delignification process has been extensively applied in pulp industries.^{106,107} In this process, 301 302 lignocellulosic biomass is mixed with distilled water, sodium chlorite, and acetic acid and stirred at 70-80 °C for 4-12 h.^{106,108-110} At different intervals, pH is controlled by adding acetic acid and 303

sodium chlorite to the mixture. Distilled water is added to the stirring mixture until the pH becomes 304 neutral. The obtained solid product as fibers mainly consists of cellulose and hemicellulose and is 305 known as holocellulose, which is then dried in the oven at 50 °C. The white color of holocellulose 306 fiber suggests that the lignin and other impurities have successfully been removed.¹⁰⁹⁻¹¹¹ 307 The alkaline treatment is used to eliminate the amorphous polymers and the rest of the lignin.¹⁰⁸⁻ 308 ¹¹⁰ During this treatment, sodium hydroxide (4-20% w/v) is mixed with holocellulose for 1-5 h, 309 and the obtained solid products are washed with distilled water until the pH becomes neutral.^{106,109} 310 It is then dried in an oven at 50 °C, and the obtained solid product mainly consists of cellulose.¹¹¹ 311 Many research groups have used these pre-treatment methods to eliminate non-cellulosic materials 312

- from lignocellulosic biomass as demonstrated in Figure 2.
- 314

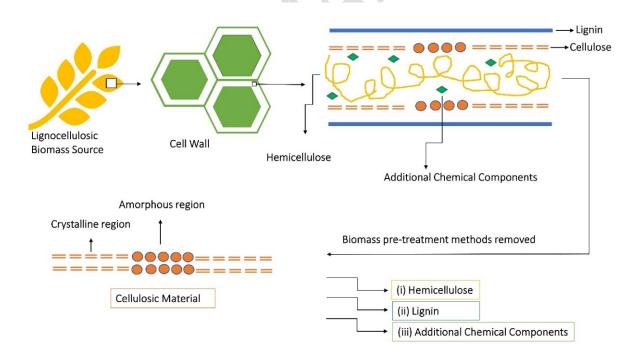


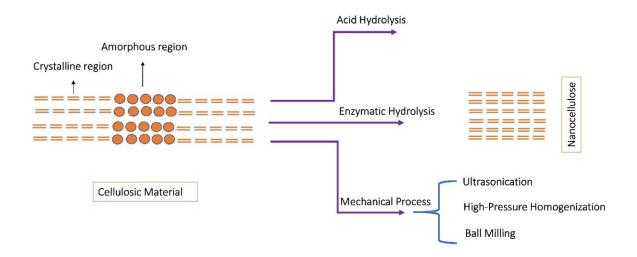


Figure 2: Schematic representation of lignocellulosic biomass pre-treatment

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318 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.^{8,38} The properties of extracted nanocellulose depend on the temperature, acid concentration, and reaction time during acid hydrolysis.^{28,38,112} Sodium hydroxide is used to neutralize the pH of the obtained products.¹¹³



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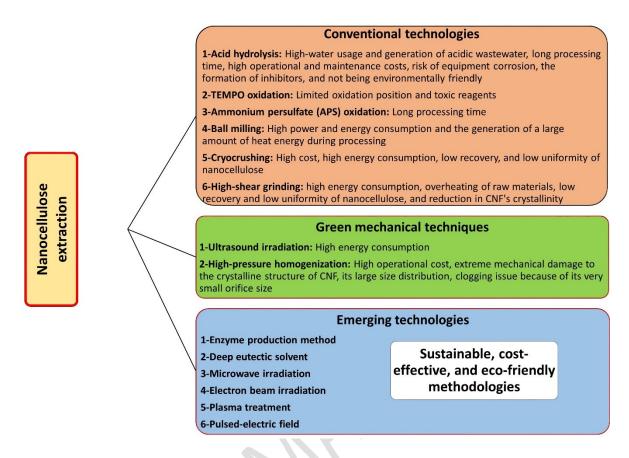
Figure 3: Schematic representation of production of nanocellulose

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In enzymatic hydrolysis, enzymes are used to digest or modify the cellulose fibers.^{114,115} 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.^{28,38} 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 332 fibers into cellulose nanofibrils.77,78,114 The mechanical process includes ultrasonication, high-333 pressure homogenization (HPH), and ball milling. During ultrasonication, the hydrodynamic force 334 of high-intensity ultrasonication is used to defibrillate cellulosic fiber.^{78,114,116} When the liquid 335 molecules are subjected to absorb ultrasonic energy, the mechanical oscillating power is generated, 336 which results in the formation, expansion, and collision of microscopic gas bubbles.^{117,118} The 337 high-pressure homogenization is carried out by passing cellulose slurry through a vessel under 338 high pressure and velocity.¹¹⁴ 339

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.^{119,120} The cellulose fibrils are broken down into smaller cellulosic materials.¹²¹⁻¹²³ However, the main drawback of this method is high energy consumption, which could be decreased by combining it with other pretreatment methods.¹¹⁴ 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.



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

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351 6. Applications of Nanocellulose

352 Nanocellulose has been an attractive material with multiple applications due to its extraordinary

353 properties (Figure 5). A detailed description of each application is given below:

354 6.1. Composite Industry

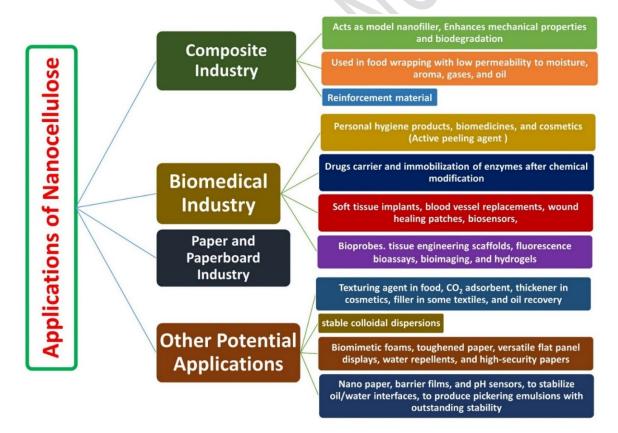
355 In recent years, the use of nanocellulose in the field of polymer reinforcement to create high-

356 performance biomaterials has become the principal subject of interest. It is due to its nano-size,

- 357 high aspect ratio, good dispersion in hydrophilic systems, higher uniformity with fewer defects,
- 358 and enhanced mechanical properties.¹²⁴ It could be applied as a reinforcement material to

359 synthesize nanocomposites with water-soluble polymer solutions to improve the mechanical360 strength and viscosity of dry composites.

One of the most interesting uses is its addition to biodegradable polymers, which not only enhances 361 the mechanical properties of the polymers but also enhances their biodegradation.^{125,126} It has been 362 a model nanofiller in many polymeric nanocomposite systems as it can improve mechanical 363 364 strength and modulus even at very low concentrations. Nanocellulose-based polymeric composites can be utilized to develop membranes, textiles, fibers, electroactive polymers, batteries, 365 supercapacitors, sensors, and actuators that utilize electromechanical responses.^{127,128} Its 366 incorporation into different materials can improve mechanical strength, optical and thermal 367 stability, and barrier properties because of its better interfacial interaction and crystallinity. 368



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Figure 5: Applications of nanocellulose in various fields

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Nanocellulose-based biodegradable nanocomposite films can also be used in food wrapping where
lower permeability to moisture, aroma, gases, and oil is required.¹²⁹

374 6.2. Biomedical Industry

Nanocellulose is also used in the field of medical sciences due to its good biocompatibility, low 375 toxicity, renewability, and outstanding physical features. Pure nanocellulose is harmless to humans 376 377 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 378 enzymes after chemical modification.^{36,83,130} Other interesting medical applications of 379 nanocellulose include soft tissue implants, blood vessel replacements, and many other useful 380 applications that are still under investigation in recent years.^{83,130} Nanocellulose-based aerogels 381 are also gaining attention in the field of biomedicine and pharmaceutical industries due to the 382 unique structure and higher surface area of nanocellulose, which improves drug bioavailability and 383 drug-loading capacity.¹²⁵ Cellulose nanomaterials have also been used in wound healing patches, 384 biosensors, bioprobes, tissue engineering scaffolds, fluorescence bioassays, bioimaging, and 385 hydrogels for medical and pharmacological applications.¹³¹ 386

387 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.¹³² 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.¹³³ 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.¹³⁴ It improves the structural properties (flexibility and transparency) of paperboard to be used for electronic devices.¹²⁶ Modified nanocellulose-based paper is transparent, optically clear, and foldable, and can be used in areas that require surfaces with anti-corrosion and selfcleaning 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.

398 6.4. Other Potential Applications

Nanocellulose can also be used as a texturing agent in food, CO₂ adsorbent, thickener in cosmetics,
filler in some textiles, oil recovery, and many other fields.^{15,135-137} 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.

Though stable dispersions in the presence of nonpolar solvents can also be achieved by using either 403 surfactants or surface chemical grafting, however, it is also possible to disperse nanocellulose in 404 nonpolar solvents without surfactant or chemical modification.¹³⁸⁻¹⁴¹ Besides good dispersion, 405 nanocellulose, in some cases, forms a percolated network within a polymer matrix, which 406 improves the features of polymeric nanocomposites. Such kinds of nanocomposites are used to 407 produce biomimetic foams, toughened paper, versatile flat panel displays, water repellents, and 408 high-security papers.^{95,133,142,143} Other distinctive applications of nanocellulose are its use in a 409 range of products such as nano paper, barrier films, and pH sensors, to stabilize oil/water 410 interfaces, and to produce Pickering emulsions with outstanding stability.^{95,144-146} 411

412 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.¹⁴⁷ 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.¹⁴⁸ However, due to the low mechanical performance (hardness, scratch
resistance) of waterborne coatings, their application in the waterborne coating industry has become
limited.¹⁴⁹⁻¹⁵²

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.^{47,151,167,168,174,175}

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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	Reference
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	64
Salvia-based waterborne polyurethane-urea (WBPUU)	Cellulose nanocrystals (CNC)	CNC addition modulates the soft and hard phase segregation	Effective CNC incorporation without agglomeration	153
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	154
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	155
PVA coating	Polyethylene glycol methacrylate (MPEG) and cellulose nanocrystals	Strong reinforcing effect with high transparency	Colloidal stability	156
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	157
Waterborne acrylate-based matrix coating	Cellulose nanocrystals	Improved barrier performance	Barrier performance	158

Waterborne acrylate formulation	CNC or cellulose	CNC-based barrier coating produced	Stable polarization	159
	nanofibrils (CNF)	had high stable polarization resistance	resistance, open circuit	
		and open circuit potential	potential	
WPU paint	Nanocellulose	Improved hardness and abrasion	Improvement in	160
		resistance, tensile strength, and	comprehensive	
		elongation at break	properties	
Photopolymerizable	Nanocellulose and	Improved hydrophobicity, thermal	Improvement in	161
siloxanemethacrylic-based	microcellulose	properties, stiffness into the neat cured	comprehensive	
resin/CNC		systems with no embrittling, surface	properties	
		hardness with reduced water sorption		
Epoxy resin-based	Nanocellulose	Improved anti-corrosion properties	Improved	162
nanocomposites	loadings		physicochemical and	
			anti-corrosion properties	
Composite of TEMPO-oxidized	TOCNs	Enhanced the mechanical properties	Improvement in	163
cellulose nanofibers (TOCNs) and			mechanical properties	
WPU-based coating				
CNC modified by a cationic	CNC was modified by	CNC improved the barrier and optical	Improvement in the	164
surfactant was added to the	a cationic surfactant	properties and agglomeration	barrier and optical	
coating system	at two loadings (1		properties	
	and 3%).			
Composite of CNFs with γ-	γ-	Demonstrated distributing uniformly,	High light transmittance	165
aminopropyltriethoxysilane (APS)	aminopropyltriethox	retained high light transmittance, and	with improved	
	ysilane	improved mechanical properties.	mechanical properties	
Dispersion and stabilization	Nanofibrillated	Improved unstabilized ZnO distribution	Improved distribution of	166
impact of zinc oxide (ZnO)	cellulose (NFC) and	and prevented sedimentation of ZnO,	unstabilized ZnO in the	
nanoparticles in waterborne wood	zinc oxide (ZnO)	improved film formation, and inhibited	coatings, film formation,	
coatings.		crack formation. NFC had a pronounced	and inhibition of crack	
		matting effect	formation	
Composite films of NFC, and	Acrylic and alkyd	Improved reinforcing effect	Improved mechanical	167
commercial acrylic and alkyd	polymeric binders		properties	
polymeric binders				
Carbon chains grafting of cellulose	Alkyl quaternary	CNC derivatives were better dispersed,	CNC dispersion and	168
nanocrystal (CNC)	ammonium bromides	conferring a higher scratch resistance	mechanical properties	
	or acryloyl chloride			

CNC was mixed in the varnishes	Varnishes	Improved wear resistance (30-40%)	Mechanical properties	150
Microfibrillated cellulose (MFC) and nanocrystalline cellulose (NCC) were added to waterborne	MFC and NCC	Lowered gloss, improved hardness, and modulus	Improvement in mechanical properties	151
Commercially available paperboards were coated with thin layers of nanocellulose	Commercially available paperboards	Improved water vapor barrier properties	Water vapor barrier properties	61
ZnO and polymer composites	CNC	Reduced destructive effects of UV light and humidity	Reduced ΔE value	176

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Nanocellulose has received tremendous attention in the sustainable biomaterial-based industry due 440 to its unique nature. Coating technology is mostly applied in the construction, furniture, 441 automobiles, military, and packaging industries.¹⁷⁷ Nanocellulose improves service life and plays 442 an essential role in its functionality. Regarding its use in coatings, several studies have been 443 published, but its use in waterborne wood coatings is still in progress, and limited research work 444 445 has been reported. Nanocellulose has improved resistance against scratching and abrasion of acrylic latex, high-solids coating, and UV-curing waterborne varnishes.^{150,164,168,169} Besides 446 developments in the field of coating, also optical properties may be affected by the addition of 447 nanocellulose to coatings.¹⁷⁸ It is reported that it can be straightforward to disperse in waterborne 448 coatings; however, chemical hydrophilization would be required for nonpolar systems such as an 449 organofunctional silane or acryloyl chloride, or alkyl quaternary ammonium bromides.^{168,169} 450

451 8. Conclusions and Future Perspectives

The main focus of this review is to overview the process of production of nanocellulose (CNCs, 452 CNFs, and BC) from lignocellulosic biomass through different processes and discuss its potential 453 applications. Several research projects have worked on the production of nanocellulose from 454 various sources of lignocellulosic biomass and it is still the main challenge to make it easily 455 456 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 457 has special morphology and geometric proportions, high specific surface area, crystallinity, liquid 458 459 crystalline behavior, rheological properties, mechanical stability, alignment and orientation, barrier properties, chemical reactivity of the surface, lack of toxicity, biocompatibility, 460 461 biodegradability, many more others special features which make it a potential candidate to 462 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 463 others. It also has the potential to replace synthetic or petrochemical-based materials to enhance 464 the environmental footprint of many of these industries. While the subject of nanocellulose has 465 been researched intensively over the last few years, still the place for new advancements exists 466 such as its production from novel sources, and its application in various fields should attract more 467 attention in future studies. Due to its cost-effectiveness and environmentally friendly nature, 468 further expansion in the limits of nanocellulose applications, especially in the field of material 469 science and engineering, would be of particular interest in the future for scientists and technologists 470 471 around the globe. Similarly, the production of nanocellulose is still expensive and requires energyintensive procedures, and could be resolved by upscaling the manufacturing and economical 472 surface modification methods. Overcoming all these obstacles may further expand nanocellulose's 473 role in enhancing material science and engineering while also promoting sustainability. 474

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476 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 477 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.; 478 479 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., 480 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., 481 482 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, 483 484 A.D., R.I., S.M.E., I.A., and S.B.

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486 "The authors declare no conflict of interest."

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488 **References**

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