

Review

# Recent Advancements in the Valorization of Agro-Industrial Food Waste for the Production of Nanocellulose

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**Abstract:** The rising climate change concerns over the excessive exploitation of non-renewable sources have necessitated the need for alternative renewable and eco-friendly resources for the production of innovative materials, achieving the targets of bioeconomy. Lignocellulosic biomass (LB) constituted by polymeric sugars and lignin is an abundantly available plant-based renewable material in the form of agro-industrial food waste and crop residues that can be exploited for the production of an array of value-added chemicals and bioproducts. Cellulose is the most abundant natural and biodegradable organic polymer on earth derived from LB, with wide scale applications in the lignocellulosic biorefineries and food industries. The negative effects of food waste from agro-industrial activities could be reduced through the recovery of cellulose from these wastes and converting it into valuable forms. However, the extraction of cellulose from LB is a difficult task owing to the recalcitrant nature of biomass that restricts the easy accessibility of cellulose for value addition. Therefore, a suitable cellulose extraction method through the fractionation of LB is necessary for a better cellulose yield. Furthermore, owing to the enormous potential of nanocellulose (NC), researchers are keenly interested in developing ecologically acceptable cellulose extraction methods. Cellulose nanofibrils and nanocrystals confer excellent mechanical properties, non-toxic characteristics and biodegradability, due to which they possess wide-scale applications in diverse industrial sectors. The current review emphasizes the potential role of cellulose extraction and NC production from agro-food waste. The different pretreatment methods for their extraction from LB are outlined. The applications of nanocellulose in different areas are also discussed. The review also highlights the recent trends, challenges and future directions in the development of cellulose and NC-based commercial products.

**Keywords:** agro-industrial food waste; lignocellulosic biomass; cellulose; nanocellulose; pretreatment; circular bioeconomy



**Citation:** Nargotra, P.; Sharma, V.; Tsai, M.-L.; Hsieh, S.-L.; Dong, C.-D.; Wang, H.-M.D.; Kuo, C.-H. Recent Advancements in the Valorization of Agro-Industrial Food Waste for the Production of Nanocellulose. *Appl. Sci.* **2023**, *13*, 6159. <https://doi.org/10.3390/app13106159>

Academic Editors: Giorgia Spigno and Andrea Bassani

Received: 27 April 2023

Revised: 10 May 2023

Accepted: 15 May 2023

Published: 17 May 2023



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## 1. Introduction

In recent years, the unrestricted use of non-renewable fuels and their endless exploitation have had a significant influence not only on the environment's ability to sustain itself, but also on public health [1,2]. As a result, research is being done on the viability of natural

biomaterials, particularly the use of biopolymers made from a variety of naturally occurring biomass feedstocks [3]. Cellulose, which is the most prevalent natural polymeric substance on the planet, can serve to generate a variety of biomaterials and compounds [4]. This cellulose can be majorly derived from the agro-industrial lignocellulosic biomass (LB), which is otherwise considered to be waste creating environment issues. The utilization of LB, especially derived from the agro-food industries for the extraction of cellulose, will not only solve the issue of food waste management, but also be a part of circular bioeconomy for the production of cellulose- and non-cellulose-based value-added biomaterials [5,6].

Out of a variety of biomaterials derived from cellulose, its most significant and promising role has emerged in the development of novel and advanced materials such as nanocellulose. Nanocellulose (NC) is a naturally occurring nanomaterial with various merits, including its nanoscale dimensions, a high surface area, renewability, high aspect ratio, distinctive shape, high strength and strong optical properties [7]. The nanometric-sized NC is broadly classified into three types, i.e., cellulose nanocrystals (CNC), cellulose nanofibrils (CNFs) and bacterial nanocellulose (BNC). Although these three forms of nanocellulose have identical chemical compositions, they have diverse physical properties, such as different particle sizes, morphologies and crystallinities [8]. Whereas CNC possess a perfect crystalline structure, the CNFs contain both crystalline and amorphous patches that can be fabricated into an entangled matrix. Due to its exceptional physical properties, mechanical strength, biocompatibility and renewable source of production, NC finds its applications in a variety of fields, including biomedical science, paper and pulp, mechanically reinforced polymer composites, antibacterial coating materials and environmental remediation.

Pretreatment is a mandatory step in the fractionation of LB and the extraction of cellulose polymer for the NC production. In addition to significantly increasing the digestibility of cellulose, an effective pretreatment should also make it easier to recover other polymers such as lignin and hemicelluloses for value addition. A number of different pretreatment techniques have been applied for increasing the efficiency of cellulose separation from lignin and other components, including chemical, physical, biological and physicochemical methods [9–12]. However, in the recent times certain low-cost pretreatment strategies such as deep eutectic solvents, hydrothermal pretreatment, ball milling, microwaves, ultrasonication, etc., have been given importance, owing to their remarkable biomass deconstruction properties and contribution towards increasing the overall economy of the bioprocess [8,13,14].

An important factor in the production of nanocellulose is the source from where cellulose is derived. Wood pulp or pure cellulose, such as microcrystalline cellulose, are the principal sources of NC. However, non-woody plant wastes such as crop waste and agro-food waste, are rich in cellulosic portions, which therefore act as starting materials for NC production. According to recent studies, nanocellulose can be derived from LB sources such as rice straw, wheat straw, coconut husks, sugarcane bagasse, pineapple leaves, corn cobs, organic kitchen waste, citrus trash, etc. [15,16]. These waste biomasses are rich in high fiber content, which makes them a particularly intriguing feedstock for the production of nanocellulose. In the previous years, many review articles have been published in the literature on different nanocellulose production strategies [17,18]. However, in the present review, the state-of-the-art green technologies for NC production from agro-industrial food and crop waste are majorly highlighted and described. The paper summarizes advancements in the methods of extraction, particularly from LB, and developments in the uses of NC-based materials. The applications of NC in the biomedical sector, wastewater treatment, food industry, nanocomposites formation and printed electronics have been presented.

## 2. Lignocellulosic Biomass and Cellulose

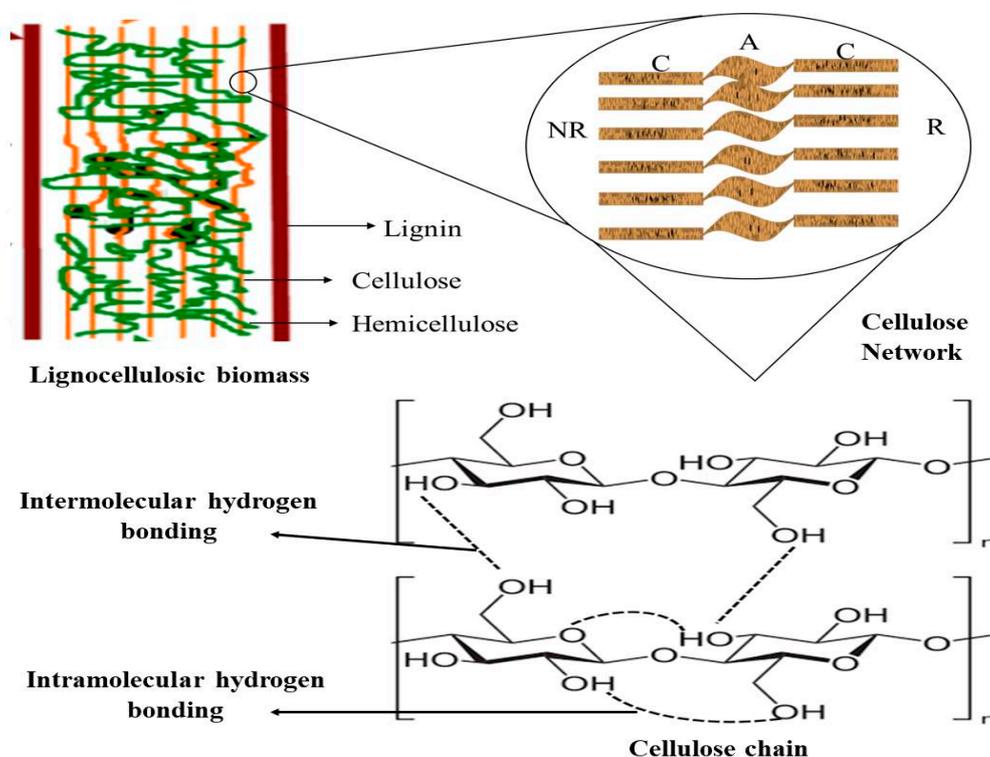
Lignocellulosic biomass, commonly known as plant biomass, is the most abundant source of sustainable carbon and the most promising raw material for the environmentally benign production of various green products, including biochemicals, bioethanol and biofuels [2]. LB is mainly that portion of a crop which is either not suitable for harvesting or

has no other end use. It completely falls under the concept of circular bioeconomy and can thereby be effectively utilized for the production of value-added products to achieve sustainable development goals [4]. The lignocellulosic materials that can be recycled include waste products from agriculture (straw, bark, stalks, leaves and bagasse), forestry (hardwood and softwood), energy crops, weeds, municipal waste, food and other sources [19]. Primarily, the structural components of LB consist of cellulose (35–50%), hemicellulose (20–35%) and lignin (15–30%), which is a polyphenolic aromatic polymer [20–22]. The primary composition of LB varies among the types of biomass, geographical area, climate, time of harvesting etc. [9]. All three components of biomass can be effectively utilized for the production of various value-added materials [1,11,23]. However, the major bottleneck in utilization of LB components for the production of different valuable products is its recalcitrant nature. The three polymers in LB (cellulose, lignin and hemicellulose) are compactly packed and interconnected with one another in a heteromatrix to variable degrees and compositions. The intrinsic packaging of these polymers, cellulose crystallinity and degree of polymerization, presence of acetyl group, hydroxyl group and syringyl-to-guaiacyl ratio in lignin are some factors that contribute to overall LB recalcitrance [2,24]. Biomass recalcitrance prevents mechanical/microbial/enzymatic action on the biomass and limits its conversion into useful materials [25–27]. Hence, reducing the biomass recalcitrance is a precondition for the efficient conversion of LB which is ideally accomplished through biomass pretreatment. Pretreatment aids in extracting/separating the desirable polymers from the complex network of three main polymers and is therefore one of the major steps in biomass conversion. After the efficient fractionation of LB, cellulose, hemicellulose and lignin polymers can be utilized for the production of various products, including biofuels, hydrogels, vanillin, organic acids and nanomaterials [1,2,4,28].

Cellulose is the most abundantly present polymer across the globe, with an estimated natural production between  $10^{10}$  and  $10^{11}$  tonnes annually, out of which only about  $6 \times 10^9$  tonnes are utilized by various industrial sectors, including the paper, textile, chemical and material industries [29]. Cellulose  $[(C_6H_{10}O_5)_n]$  is made up of repeating  $\beta$ -D-glucopyranose molecules linked together by  $\beta$ -(1,4)-glycosidic bonds, and constitutes the highly hard crystalline portion of the plant cell wall. The OH groups in the C<sub>4</sub> and C<sub>1</sub> carbon atoms of the glucose units form glycosidic bonds that covalently bind to form several hundred to thousand repeating units, which are linear polysaccharides [30,31]. Every glucose unit in cellulose is rotated 180 degrees in relation to its neighbors. Hydrogen bonds can be formed between the three free hydroxyl groups (OH) at the C-2, C-3 and C-6 atoms, which hold a crucial role in the crystalline packing (Figure 1). The solubility and other physical attributes of cellulose are controlled by intramolecular and intermolecular hydrogen bonding [3]. Irrespective of the type of LB, a polymeric chain of  $\beta$ -(1-4)-linkage and D-glucose commonly aggregates via Van der Waals interactions and inter/intra molecular hydrogen bonds in nanosized, thread-like assemblies, termed as microfibrils. These microfibrils are further packed into fibers and are arranged in a planar sheet to form a structural framework of the fiber cell wall [8,32]. The cross-sectional dimensions of a congregated cellulose chain (microfibrils) vary from 2 to 20 nm during biosynthesis on the basis of the cellulose source. The cellulose fibrils contain two types of regions/areas: one is a crystalline region where the cellulose chains are organized in a highly ordered form, and another is an amorphous-like region where they are disordered. These crystalline regions provide strength and stiffness per unit of weight and are interspersed with amorphous areas [19,33].

The crystalline cellulose exhibits polymorphism and therefore occurs in various structural confirmations due to the variation in the orientation of the glucose units and spatial hydrogen bond configuration within the network. Consequently, it results in the formation of four primary allomorphic forms of cellulose known, i.e., cellulose types I, II, III and IV. The cellulosic allomorphs can change depending on the LB source of cellulose and the pretreatment approach applied to separate the cellulose from the biomass [30,31]. The most prevalent allomorph found in nature is cellulose type I, or native cellulose, which is

characterized by a parallel arrangement of hydrogen bonding networks. Cellulose type I is dissolved in an acid or alkaline solution, or in a solvent, that subsequently undergoes chemical regeneration to become cellulose type II. These chemical regenerations lead to various arrangements of antiparallel packing of the hydrogen-bond network in cellulose type II. The supramolecular structure of the biopolymer is fundamentally changed irreversibly when cellulose type I is converted to type II, where the hydroxymethyl groups experience conformational changes and lack an intramolecular hydrogen bond along the chain axis [34]. Ammonia treatment of cellulose types I and II results in the production of cellulose type III, which can then be modified into cellulose type IV by heating in glycerol for 20 min at 260 °C [35]. Among the biopolymers that make up lignocellulosic fibers, the degree of polymerization is the highest in cellulose and depending on the cellulose source, it can range up to 20,000 units or more with a value of around 10,000 in wood [33,36]. Different types of cellulosic fibers can be produced owing to the abundance and excellent benefits of agro-industrial biomass, including light weight, economy, biodegradability and pervasiveness [37].



C= crystalline cellulose; A= amorphous cellulose; NR= non-reducing end; R= reducing end

Figure 1. Chemical structure of cellulose from lignocellulosic biomass.

### 3. Nanocellulose

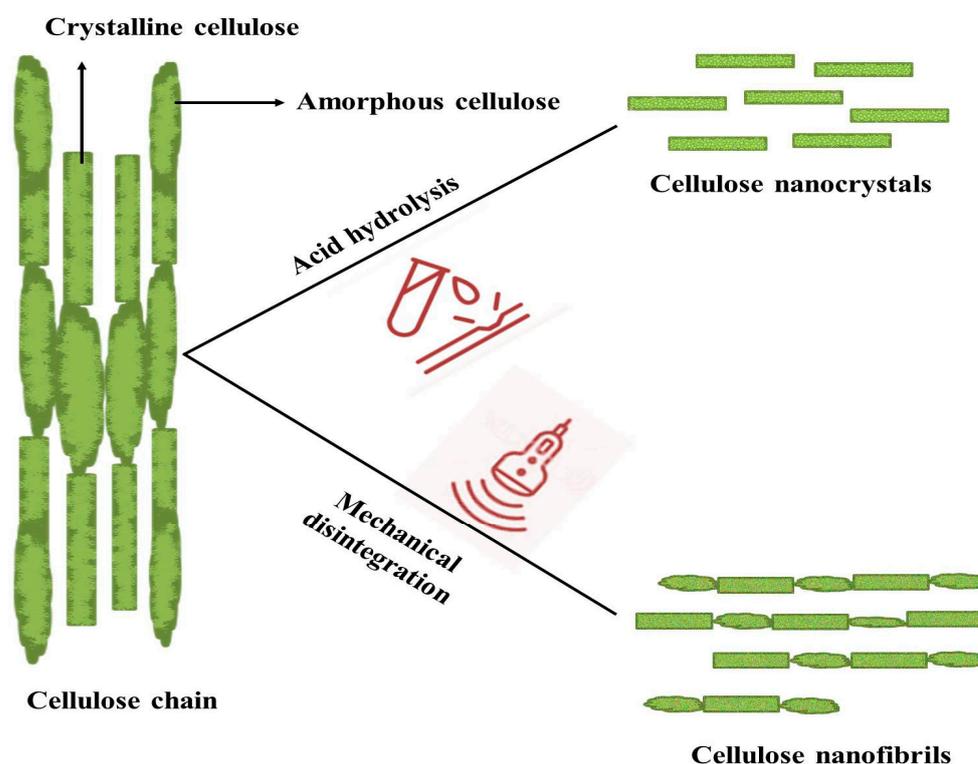
In recent years, aside from the production of biofuel and other chemicals, the global research focus has switched to the synthesis of nanomaterials such as nanocellulose (NC), utilizing cellulose from LB. NCs are cellulose fibrils or crystallites that have at least one dimension falling within the region of the nanoscale. Generally, the aspect ratio of NC is high, with a length of up to several microns and lateral dimensions of lower than 100 nm [35]. NC made from different sources of cellulose may differ in the amount of cellulose present, degree of crystallinity, and most notably, the size of the crystalline domains [31]. NC possesses excellent properties, such as special morphology and geometrical dimensions, crystallinity, mechanical strength, high specific surface area, thermal stability, rheological properties, stiffness, alignment and orientation, barrier properties, light weight, non-toxicity,

biodegradability, low density (around  $1.6 \text{ g/cm}^3$ ), biocompatibility and surface chemical reactivity. In particular, it has a higher stiffness than Kevlar fiber, with an elastic modulus of up to 220 GPa. Owing to these remarkable attributes, NC demonstrates a significant potential for a wide range of applications, providing new opportunities for the production of novel value-added materials, including food packaging, polymer and paper reinforcement, additives to composites emulsions, barrier films, water treatment and within the health care sector [8,19,35]. However, it has been reported in some studies that nanocellulose possesses low thermal stability, which restricts their application and the conditions in which their nanocomposites can be produced at high temperatures. This problem is especially serious for cellulose nanocrystals that have been hydrolyzed in sulfuric acid, since it has much less heat stability than the original starting material or other types of nanocellulose [38].

The presence of a large number of hydroxyl groups aids in the functionalization of NC and the tuning of its various surface properties. This characteristic of NC paves the way for their application in different areas. The source of cellulose, its isolation process, as well as pre- and post-treatment greatly impacts the shape, architecture, structure, size and other properties of NC. Additionally, it is feasible to model the properties of the NC for a particular application based on the source from which the cellulose is derived, the pre-treatment and the conversion methods selected, bringing up a wide range of opportunities [19,35]. The three major types of NC, i.e., cellulose nanocrystals (CNCs), cellulose nanofibrils (CNFs) and bacterial nanocellulose (BNC), are discussed in the below sections. Despite having identical chemical compositions, the various types differ from one another in terms of shape, particle size, crystallinity and other characteristics, as a result of variation in the method of isolation and source of cellulose.

### 3.1. Cellulose Nanocrystals (CNCs)

CNCs are also known as nanocrystalline cellulose, crystallites, whiskers, rod-like cellulose microcrystals. CNCs contain 100% cellulose which is organized in a crystalline fashion and is highly crystalline (54–88%). The diameter of CNCs is not dependent on the source of cellulose and mostly falls in the range of 5–70 nm. However, the length of CNCs is source-dependent, i.e., for CNCs extracted from cellulose of plant biomass, it lies between 100 and 250 nm, and 100 nm to several micrometers if the cellulose sources are tunicates, algae or bacteria. The inconsistency in the length of CNCs from different cellulose sources is attributed to the amount of crystalline cellulose present in the source, as the cellulose from tunicates and bacteria is highly crystalline and contains less amorphous regions, the length of CNCs produced from these sources is greater. Additionally, the length-to-width ratio typically falls between 10 and 100 [28,31,33]. Acid hydrolysis is the most common and efficient method for creating nanocrystalline cellulose (Figure 2). During the hydrolysis process, the amorphous region is highly disrupted owing to its high susceptibility toward acids as compared to crystalline regions. This results in the production of rod-shaped cellulose nanocrystals with variable geometrical dimensions, large surface area ( $\sim 400 \text{ m}^2/\text{g}$ ) and high crystallinity [39,40]. In comparison to other varieties of nanocellulose, the CNCs stand out because of their inherent qualities, which include high tensile strength (7500 MPa), rigid structure, amphiphilic nature, optical properties, redispersion from dried powder, chemical purity and a high aspect ratio ( $\sim 70$ ) [7,36].



**Figure 2.** Production of cellulose nanocrystals and cellulose nanofibrils from a cellulose chain.

### 3.2. Cellulose Nanofibril (CNF)

CNFs contain both a crystalline region and an amorphous region of cellulose in an alternate fashion, and thus represent 100% cellulose. CNFs are also named as cellulose nanofibers, nanofibrillated cellulose (NFC) and microfibrillated cellulose (MFC). These are made up of entangled bundles of flexible, long nanofibrils that can be extracted from cellulose through mechanical disruption (homogenization, microfluidization, grinding, cryo-crushing, high-intensity ultrasonication), during which the cellulose fibrils are cleaved within the longitudinal axis (Figure 2). The long thread-like fibrils present in CNFs are 1–100 nm in diameter and 500–2000 nm in length. It is expected that CNFs have axial tensile strengths and Young's modulus of approximately 3 GPa and 136 GPa, respectively. Furthermore, CNFs exhibit thixotropic and shear-thinning behavior when they form gels in water [19,41,42]. Comparing CNFs with CNCs, the former has a longer length, a higher aspect ratio (length-to-diameter), a larger surface area and a higher concentration of hydroxyl groups, which makes it more easily accessible for surface modifications [43]. Due to their extremely high aspect ratios and fibrillar shape, CNFs in particular exhibit exceptional potential in a variety of applications, such as flexible electronics, high performance polymer composites, separation media and adhesive binders [44]. While mechanical disintegration is usually applied to release the nanofibers, the procedure, being very energy-consuming, is believed to be a significant hurdle to the effective production of CNFs. Nevertheless, introduction of various pretreatment techniques added before the mechanical processes, as one of the pre-requisites with the goal of lowering energy consumption and making the surface hydrophobic, may aid in a successful commercial production of CNF [19,41,44].

### 3.3. Bacterial Nanocellulose (BNC)

Contrary to CNC and CNF which are produced from cellulose extracted from plant biomass, BNC, as the name suggests, is nanocellulose produced by bacteria. It is made from the fermentation of glucose or other carbohydrate feedstocks by bacterial processes, and is also known as microbial cellulose or bio cellulose [31,35]. It is mainly produced by *Gluconacetobacter xylinus* (*Acetobacter xylinum*) [45]; however, genera such as *Acetobacter*,

*Achromobacter*, *Bacillus*, *Sarcina*, *Aerobacter*, *Agrobacterium*, *Escherichia*, *Azotobacter*, *Rhizobium*, *Enterobacter*, *Klebsiella* and *Salmonella* also produce cellulose. The synthesis of bacterial cellulose typically takes place in four stages: in the first stage, monosaccharides are activated through sugar nucleotide development; in the second stage, polymerization occurs, which leads to the formation of assembly of repeating sugar units; the third stage is characterized by the in-time addition of any acyl group if present; and in the last stage, extracellular secretion of BNC in the form of three-dimensional microfibril and nanofibril network takes place [46]. Enzyme cellulose synthase plays the role in the synthesis of BNC and the BNC produced by *Gluconacetobacter xylinus* that possesses a chemical structure identical to plant cellulose. Since BNC is produced from bacteria and not from plants, it does not contain impurities such as lignin, pectin, hemicellulose, wax, etc. [17,46–48]. BNC is also traditionally used to produce a common dessert in Southeast Asia, called ‘Nata de coco’, through the fermentation of coconut water. Currently, many agricultural wastes have been reported that can be fermented for the production of bacterial cellulose, such as orange peel [49] and pineapple peel [50]. BNC has a twisted ribbon topology and usually has a diameter of 20 to 100 nm, length up to 100  $\mu\text{m}$  and a large surface area per unit. The morphology of BNC is dependent on factors such as cultivation mode and cultural parameters [17,35]. A high porosity, high elastic modulus, high degree of polymerization (up to 8000), high water-holding capacity and high crystallinity (up to 84–89%) are only a few of the distinctive physical and mechanical attributes of BNC. BNC may possess a promising potential application in various fields such as biomedical applications, electronic paper, fuel cell membranes and reinforcement in nanocomposites [41,43]. The properties of CNC, CNF and BNC are summarized in Table 1.

**Table 1.** Summary of various properties of different types of nanocellulose.

Properties	Cellulose Nanocrystals	Cellulose Nanofibrils	Bacterial Nanocellulose	Reference
Diameter	5–70 nm	1–100 nm	20 to 100 nm	[33,35,41]
Length	100–250 nm (plant cellulose) and 100 nm–several micrometres (cellulose sources from tunicates or algae)	500–2000 nm; can reach up to $\mu\text{m}$	up to 100 $\mu\text{m}$	[17,33,41]
Cellulose	Crystalline	Crystalline and amorphous	Crystalline and amorphous	[17,33,41]
Tensile strength	~7500 MPa	~3 GPa	200–300 MPa	[33,42,48]
Young’s modulus	~110 GPa	~136 GPa	15–35 GPa	[42,48]
Impurities	Presence of lignin, hemicellulose, pectin and wax	Presence of lignin, hemicellulose, pectin and wax	No impurities	[17,46–48]

#### 4. Technologies for Nanocellulose Production

The production of nanocellulose from agro-industrial food residues or from any other lignocellulosic biomass is broadly divided into two steps: (1) the extraction of cellulose from lignocellulosic biomass and removal of undesirable non-cellulosic components, and (2) the preparation of nanocellulose from extracted cellulose [8]. The isolation of cellulose from LB is usually achieved through the pretreatment process, which has become a prerequisite in any biorefinery using LB as a raw material for the production of useful products.

##### 4.1. From Lignocellulosic Biomass to Cellulose

The inevitable step in the conversion of LB into any value-added product is its pretreatment owing to the intricacy of the lignocellulosic structure, as mentioned in Section 2. An efficient pretreatment regimen may help in breaking down the chemical and physical barriers that prevent the fractionation of LB without producing any inhibitors [11,51]. The lignocellulosic structure is completely disintegrated as a result of the pretreatment of the biomass, which encourages an extensive cleaving of chemical bonds connecting the polymers within the lignin–carbohydrate complex (LCC). This results in an increased accessibility of enzymes to the polysaccharides [11]. Chiefly, an ideal pretreatment method

should be: (1) suitable for various kinds of LB, (2) cost and energy effective, (3) not produce toxic or inhibitory compounds, (4) produce highly digestible pretreated solid and (5) have high reaction efficiency [52]. The pretreatment of biomass can be achieved through various physical (ultrasound, microwave), chemical (ionic liquids, deep eutectic solvents, acids, alkali), physicochemical and biological techniques [20,51,53,54]. Ideally, combinatorial pretreatment approaches are also necessary to improve the porosity of the fiber matrices for pretreatment chemicals to permeate more easily, as well as remove any undesirable non-cellulosic components without changing the crystalline components of the biopolymer [9].

#### 4.1.1. Physical Pretreatment of Lignocellulosic Biomass

##### Ultrasound Pretreatment

Ultrasound pretreatment, a green non-thermal physical method, has recently emerged as the pacesetter, and is widely implemented and studied by the scientific fraternity for LB pretreatment [10,55]. Ultrasonic waves are longitudinal sound waves with frequencies above 20 MHz, which is higher than the hearing limit of a human. Essentially, ultrasound pretreatment is based on cavitation which breaks the  $\beta$ -O-4 and  $\alpha$ -O-4 linkages of lignin and separates lignin and polysaccharides leading to the formation of cavitation bubbles [56]. As these bubbles reach a threshold size, they violently collapse, increasing temperature and pressure by 2000–5000 K and 1800 atm, respectively, at a small temporal and spatial scale, which is called shockwaves/hotspots. These shockwaves bring the alterations in the morphology of the biomass by generating turbulence, and cause shear and shock [56,57]. Ultrasonication results in both physical and chemical changes in the biomass, including morphological disruptions, reduction in the degree of polymerization and an increase in specific surface area. The cavitation also induces strong microturbulence, which results in the micromixing in the medium and eliminates mass transfer restrictions [30,58]. Ultrasound treatment has been used alone or in combination with alkali, ionic liquids or deep eutectic solvents for the pretreatment of biomass [9,54,55,59]. In a previous study, pretreatment of brewer's spent grain using ultrasound was executed for releasing fermentable sugars [59]. The pretreatment process was optimized using response surface technology that resulted in  $325 \pm 6$  mg/g biomass of reducing sugar yield, which was 2.1-fold more than the untreated brewer's spent grain. Ultrasound-assisted NaOH pretreatment of groundnut shells, coconut coir and pistachio shells enhanced the delignification to 71.1, 89.5 and 78.9% from 41.8, 45.9 and 38% for groundnut shells, coconut coir and pistachio shells, respectively [59]. Ultrasound technology has given a variety of bioconversion processes the much-needed boost. Ultrasonication has also been employed for NC production. The subsequent homogenization and high-intensity ultrasonication processes were employed for the preparation of CNF from cellulose extracted from rice straw. The CNFs had good thermostability with a diameter between 6 and 20 nm, and an aspect ratio of about 177 when ultrasonication was used at a 500 W power for 40 min [60].

##### Microwave Pretreatment

Microwave (MW) pretreatment is a non-conventional thermal pretreatment for the fractionation of LB. Microwaves are non-ionizing electromagnetic, low-frequency waves, with a spectrum between 300 and 300,000 MHz, and a wavelength in the range of 1 mm to 1 m [56,61]. As microwaves induce dipoles to align with the oscillating electric field, they produce thermal effects that break hydrogen bonds, which results in the disintegration of cellulose chains and thereby the breakdown of cellulose structure [62]. Traditional heating, conduction, convection and radiation cause the heat transfer from the outside of the material to the inside, whereas microwaves directly convert electromagnetic energy into heat, which causes heating at the molecular level. This results in the uniform distribution of heat to the component, and prevents overheating [7]. During the MW pretreatment, the specific polar regions inside the biomass are created, called the internal 'hotspots' that endorse the LB fractionation and swelling.

To increase the effectiveness of the microwave pretreatment, it can be combined with various chemicals, such as alkali, which are used for biomass pretreatment [14]. In a study, brewer's spent grain was subjected to microwave-assisted alkali pretreatment. The pretreatment resulted in delignification, as the lignin content was reduced to  $2.77 \pm 0.9$  g/100 g biomass (acid-soluble lignin) and  $9.09 \pm 0.65$  g/100 g (acid-insoluble lignin) from  $7.12 \pm 0.4$  g/100 g and  $23.36 \pm 1.2$  g/100 g, respectively, for the untreated biomass [63]. Lignin and xylan removal of 42.81% and 74.73%, respectively, was achieved when *Pinus bungeana* was pretreated with microwave and deep eutectic solvent combined [64]. Microwave pretreatment was used for the delignification of corn cobs prior to nanocellulose production. At the optimal conditions of 180 W power, 12.86% sodium chlorite and 16 min of reaction time, delignification of approximately 97.31% was accomplished [65].

#### 4.1.2. Chemical Pretreatment of Lignocellulosic Biomass

##### Acid Pretreatment

LB pretreatment with acid has extensively been studied by different researchers over the years. Acid pretreatment usually solubilizes hemicellulose and a little amount of lignin by breaking glycosidic bonds and increasing the accessibility of enzymes to cellulose. The pretreatment with acid can be carried out in two ways, either concentrated acid (30–70%) at a low temperature (<50 °C) or dilute acid (0.1%) at a high temperature (>200 °C). However, concentrated acids are not frequently used for pretreatment owing to their limitation of corroding the vessel, toxicity and production of inhibitors. Various types of acids are used for the pretreatment, including acetic acid, ascorbic acid, oxalic acid, phosphoric acid and maleic acid. However, hydrochloric acid and sulfuric acid are most commonly used [66–68]. A study reported the ascorbic acid (AsA)-assisted dilute acid pretreatment of wheat straw, corn stover and corncob. The pretreatment resulted in enhanced cellulose recovery of 74.87%, 75.02% and 89.47% for wheat straw, corn stover and corncob, respectively [69]. Pretreatment of loblolly pine using dilute acid (H<sub>2</sub>SO<sub>4</sub>) effectively removed 80% (*w/w*) hemicellulose and improved the delignification rate by 17%, which resulted in the production of 97% pure cellulose [70].

##### Alkali Pretreatment

Alkali pretreatment mostly aids in the removal of lignin and solubilizes hemicellulose to a little extent. The alkali pretreatment is usually used for the LB which has a high lignin content [71,72]. Alkali pretreatment disrupts the lignin carbohydrate complex by hydrolyzing ester, aryl-ether, alkyl-aryl linkages via the process of saponification and salvation reactions. The acetyl and uronic acid groups in hemicellulose are also removed using alkalis. Generally, sodium hydroxide, potassium hydroxide and calcium hydroxide are used in the biomass pretreatment for delignification [57,71]. Shahabazuddin et al. reported the pretreatment of rice husk using a thermal-assisted alkaline approach. The pretreatment was effective in lignin removal (54%, *w/w*), mildly solubilized hemicellulose (32.65–51.65%, *w/w*) and increased the concentration of cellulose (51.65%, *w/w*) [73]. In another study, water hyacinth was subjected to NaOH pretreatment (5%), which was followed by bleaching treatment using 2% acidified NaClO<sub>2</sub> for nanocellulose preparation. The pretreatment led to an enhanced cellulosic content (76.57%), and decreased non-cellulosic components (lignin and hemicellulose) [74].

##### Ionic Liquid Pretreatment

Ionic liquid (IL) pretreatment of LB is a greener pretreatment strategy, which has gained a lot of attention over the last decade. ILs are salts of organic cation and inorganic/organic anions that are liquid at room temperature (<100 °C). They possess various green properties such as a low vapor pressure, non-flammability, non-corrosive, high thermal and chemical stability [75]. The mechanism of IL action is based on the dissolution of cellulose in biomass by IL [76]. The significant inter- and intramolecular hydrogen bonding

in the cellulose is broken down as a result of the hydrogen bond that forms between the anionic component of the IL and the hydroxyl group proton of the cellulose, which leads to the cellulose dissolution and disruption of the complex network of LB [26,75,77]. ILs are also known as “designer solvents”, as they can be designed with tunable properties by altering the branching, cation or anion substituent and length of alkyl chain of the cation according to the biomass to be pretreated [19,77]. ILs can be used alone or in combination with other physical/chemical pretreatment approaches for biomass pretreatment. Pretreatment of herb residues using 1-butyl-3-methylimidazolium chloride ([Bmim]Cl), assisted by aromatic acid (p-toluenesulfonic acid, TsOH), resulted in an increased cellulose recovery yield (96.4%) and lignin removal rate (79.9%) under optimal conditions of 79% IL, 1.0% TsOH and 20% H<sub>2</sub>O at 130 °C for 2 h [78]. Bamboo biomass was subjected to ultrasound-assisted 1-ethyl-3-methylimidazolium acetate ([Emim][OAc]) pretreatment, which resulted in 69.3% and 14.9% yield of cellulose-rich and lignin-rich materials, respectively [79]. ILs have been also used for the preparation of NC from extracted cellulose. A study reported the use of 1-butyl-3-methylimidazolium chloride (BmimCl) for the preparation of cellulose nano-fibrils from sugarcane bagasse. The diameter of the prepared cellulose nano-fibrils was between 4 and 35 nm [80].

#### Deep Eutectic Solvent Pretreatment

Deep eutectic solvents (DES) are the viable replacements of ILs, as they overcome the limitations of ILs such as high cost, toxicity and non-biodegradability, among others. DESs are made by mixing a hydrogen bond acceptor (HBA) and a hydrogen bond donor (HBD) in particular molar ratios in solid-phase, to form liquid phase eutectic mixtures at a moderate temperature (60–80 °C) and ambient pressure conditions [2,81]. The hydrogen bond (H-bond) between HBA and HBD promotes charge delocalization, resulting in a DES with a melting point lower than that of its individual components. They may also be regarded as eutectic mixtures of Lewis or Bronsted acids and bases that form a solution. A stronger H-bond causes a greater melting point decline [82]. DES pretreatment has been executed on various LBs for cellulose extraction and lignin removal. Pretreatment of corncob with DES choline chloride/ethanolamine led to the delignification of 83.12% [83]. DES choline chloride/formic acid was used for the pretreatment of corn stover, which resulted in the lignin removal of 23.8% with 47.9 ± 0.3% cellulose [84]. DES pretreatment has also been used for cellulose nanofibril preparation from ramie fibers. Pretreatment with DES choline chloride–oxalic acid dihydrate for 4 h at 100 °C resulted in the production of cellulose nanofibrils with 90.31% pure glucan, and a width of 14.29 nm and crystallinity at 79.17% [85].

#### 4.1.3. Physicochemical Pretreatment of Lignocellulosic Biomass

Physicochemical pretreatment approaches, such as steam explosion, hydrothermal treatment, etc., are more effective and environmentally friendly than traditional physical and chemical pretreatment strategies. Steam explosion (SE), also known as auto-hydrolysis, is the most extensively explored and studied physicochemical pretreatment technique for biomass disintegration. A sudden pressure change during the steam explosion pretreatment generates an explosive disintegration in the biomass and may physically damage the structure of fibers. During this pretreatment, LB is exposed to steam of very high temperature for a residence time of a few seconds to a few minutes, followed by an abrupt release of pressure, which results in the hydrolysis of the acetyl group present in hemicellulose, and ultimately the breakdown of components of the cell wall [86]. Another widely used physicochemical technique is ammonia fiber explosion, which involves the use of liquid ammonia at a temperature and pressure between 60 and 100 °C and 1.7 and 2.1 MPa, respectively, for a small time period (5–60 min). Ammonia can expand suddenly in conditions of high pressure and temperature, which contributes to the swelling of biomass, increase in surface area, hemicellulose solubilization and physical breakdown of the biomass structure [87,88].

Steam explosion with dilute sulfuric acid was used for the pretreatment of *Miscanthus × giganteus*, poplar and wheat straw. The results revealed that, irrespective of the biomass, approximately 90% of the hemicellulose fraction removed from the pretreated substrate was achieved [89]. Ammonia fiber expansion pretreatment of oil palm empty fruit bunch resulted in a reduced lignin content,  $16.84 \pm 0.95\%$  *w/w*, as compared to an untreated biomass ( $23.38 \pm 0.88\%$  *w/w*) [90]. In another work by Yeganeh et al., the effects of hydrothermal pretreatment and depressurization (DP) cycles were investigated on cellulose nanofiber from garlic skin waste biomass that improved the defibrillation, and subsequently affected the thermal stability, yield and crystallinity of nanofibers [13].

#### 4.1.4. Biological Pretreatment of Lignocellulosic Biomass

The biological pretreatment of the LB involves the use of microorganisms such as fungi and bacteria, which are allowed to grow on the biomass to be pretreated or the enzymes produced by such microbes for the degradation of lignocellulose. Enzymes play a crucial role in the bioprocessing of organic feedstocks into simpler products [91]. The biological pretreatment is a green approach which does not involve the use of any chemical, requires less energy and does not produce inhibitory or toxic by-products [92]. Various fungi (white rot, brown rot and soft rot), as well as bacterial genera (*Acetobacter*, *Alcaligenes*, *Sarcina* and *Pseudomonas*), are used for biological pretreatment. These microbes produce cellulose-degrading (cellulase, lytic polysaccharide monooxygenases and cellobiose dehydrogenase) and hemicellulose-degrading (endoxylanase, arabinofuranosidase,  $\beta$ -D-xylosidase and esterase) and lignin degrading (laccase, manganese peroxidase, lignin peroxidase,  $\beta$ -etherase and dye-decolorizing peroxidase) enzymes for the breakdown of different components of LB [4,91]. In a study, six white-rot fungi (*Trametes versicolor*, *Pleurotus ostreatus*, *Phanerochaete chrysosporium*, *Corioloropsis gallica*, *Pleurotus sajor-caju* and *Lentinula edodes*) were used for the pretreatment of corn stover. *Pleurotus sajor-caju* resulted in the highest delignification of 38.29% after 30 days of pretreatment [93].

A lignin degradation of 28.55% was achieved from tobacco straw pretreated by using *Bacillus amyloliquefaciens* SL-7 after degradation of 15 days [94]. Even though biological pretreatment offers the advantages of being environment friendly, it is very time consuming and laborious, and also involves a lot of crucial steps prior to the pretreatment such as sterilization, growth of desirable microbes without contamination, maintaining optimal cultural and environmental conditions, etc. All these factors limit the application of biological treatment on a larger scale. Therefore, to address such issues, more research and development involving cutting-edge molecular approaches are required.

#### 4.2. Production of Nanocellulose from Cellulose

The production of NC from the cellulose extracted from agro-industrial residues can mainly be achieved through chemical and mechanical approaches. To date, numerous technologies have been reported for the production of NC; however, the type of technology to be employed depends on the type of NC to be produced, i.e., CNC or CNF, since the treatment procedure has a significant impact on the type and characteristic of the NC. The various NC preparation technologies are briefly discussed in the current section.

##### 4.2.1. Acid Hydrolysis

The simplest and traditional chemical process for creating nanocellulose from cellulosic sources is acid hydrolysis, which is widely used for the production of CNCs. In acid hydrolysis, the disordered amorphous regions are digested owing to their more susceptibility to acids, as compared to the crystalline regions, leading to the high crystallinity of CNCs. Strong acids such as sulfuric acid ( $H_2SO_4$ ), hydrobromic acid (HBr) and hydrochloric acid (HCl) are usually employed to produce highly crystalline and rod-shaped CNCs. Weak acids such as lactic acid and acetic acid have also been reported for the production of CNCs; nevertheless, due to their low dissociation constant, the protonated ions ineffectively remove amorphous regions, resulting in a lower crystallinity and fibrous

structure. This limitation of weak acids can be overcome by combining the process with mechanical disintegration to produce a good-quality NC [31]. Among strong acids, sulfuric acid hydrolysis is the most extensively used method, during which the esterification of the hydroxyl groups on the surface of cellulose occurs, resulting in the large number of negatively charged sulphate groups (sulphate half-ester groups,  $\text{OSO}_3^-$ ) on the surface of CNCs. These anionic groups promote the dispersion of CNCs in water by forming and covering the CNCs, and prevent their aggregation in water. However, CNCs produced using sulfuric acid have moderate thermal stability, which can be resolved by neutralizing CNCs with sodium hydroxide [95,96]. The morphology, crystallinity, size, thermal stability and surface charge of CNCs are highly influenced by the type of LB, acid type, concentration, reaction time and temperature [97]. However, this process has a number of drawbacks, including high water usage, the generation of acidic effluent, a lengthy processing period, high operational and maintenance expenses, equipment corrosion, the formation of inhibitors and malignancy to the environment [30]. Sukyai et al. produced CNCs from sugarcane bagasse using 65 wt%  $\text{H}_2\text{SO}_4$  at 60 °C for 60 min under sonication. The length and diameter of the produced CNCs was found to be 200–300 nm and 20–40 nm, respectively [98].

#### 4.2.2. TEMPO-Mediated Oxidation

Another chemical method which is used for the production of NC is 2,2,6,6-tetramethyl piperidine-1-oxyl (TEMPO,  $(\text{CH}_2)_3(\text{CMe}_2)_2\text{NO}$ )-mediated oxidation, and is usually coupled with a mechanical disintegration method. Alkaline conditions (pH between 9 and 11) are typically used for TEMPO-mediated oxidation treatment, along with bleaching agents such as  $\text{NaClO}$ , and catalysts such as sodium bromide ( $\text{NaBr}$ ) and the oxidation initiates when  $\text{NaClO}$  is added. During the process, the oxidation of primary hydroxyl groups of C6 carbon of native cellulose takes place, which get converted into carboxylate groups, causing the repulsion between the ionized carboxylate groups and separating the cellulose fibrils to form NCs. The process also inhibits the aggregation ability of the NCs and increases the water dispersion of cellulose [18,99,100]. Using the TEMPO/ $\text{NaBr}$ / $\text{NaClO}$  system to hydrolyze lemon seeds in an aqueous solution at pH 10, followed by centrifugation, dialysis and sonication, a CNC yield of 52.01% was obtained [101]. Although the TEMPO hydrolysis technique is employed for the production of NC, it has certain drawbacks, including hazardous and toxic chemicals and a narrow range of the oxidation position [101].

#### 4.2.3. High-Pressure Homogenization

High-pressure homogenization is the most extensively used method for the preparation of CNFs. It is based on the fundamental principle of passing the cellulose slurry (0.5–1%) constantly through the small flow orifice of the homogenization valve under high pressure (>150 MPa) and velocity. This results in the generation of shear and impact forces, which break down the cellulose microfibrils into nanofibrils size by decreasing the cellulose fibrillation [99]. There are few disadvantages associated with this method such as clogging, energy intensity, disintegration of the cellulose fibers and the disruption of the crystalline structure [102]. High-pressure homogenization treatment was used for the production of NC from okara. The function properties of NC such as thickening, gelling, and thermal stability were improved by the high-pressure homogenization [103].

#### 4.2.4. Ball Milling

Another method which is used for the preparation of NC is ball milling, also sometimes known as solid-state synthesis. It causes the defibrillation of cellulose fibers with the help of a planetary ball. In this process, cellulose dispersion is put in a hollow cylindrical container which is partially packed with balls. The cylindrical container rotates around its axis, which results in the generation of high shearing forces due to the centrifugal force from the rotating container. These forces cause collision among the balls, and friction between the balls and the container. This collision and friction lead to the fractionation of cellulose fibers,

reduction in cellulose length and the degree of polymerization, and weakens hydrogen bonds to yield CNF [104,105]. For the synthesis of nanocellulose using this method, milling in a wet condition is preferred to preserve the fibrous state and avoid defibrillation to an amorphous form. The characteristics of NC produced through ball milling majorly depends on the number and size of the balls, the milling speed, the state of milling (dry or wet state), the weight ratio between the balls and cellulose, and milling time [35,106]. Some of the drawbacks of ball milling include the high usage of energy and power, as well as the production of a lot of heat during processing [30].

#### 4.2.5. Cryo-Crushing

Cryo-crushing is a different mechanical method in which the cellulose is released from its frozen state to form NC. Briefly, in this procedure cellulose is suspended in water and then subjected to liquid nitrogen, which results in the freezing of cellulose. This frozen cellulose is then pulverized by using mortar and pestle. The pressure exerted by crushing frozen cellulose fibrils generates high-impact forces, which results in the rupturing of the cell wall and the formation of nanocellulose [41,106]. The high cost, low recovery, inconsistency in NC uniformity and high energy requirement are few of the shortcomings of this process [35]. Nanocellulose preparation using chemical-assisted cryo-crushing was executed within wheat straw and soy hulls. The study revealed that the crystallinity of the nanofibers was enhanced 35% and 16% for the wheat straw nanofibers and soy hull nanofibers, respectively. Moreover, the nanofibers obtained from the wheat straw had a diameter in the range of 10–80 nm and lengths at a few thousand nanometers [107].

### 5. Applications of Nanocellulose

The future development relies on the progressive use of renewable materials by the upcoming sustainable technologies to create novel products with superior environment-beneficial characteristics. Nanocellulose can be processed chemically and mechanically into a variety of bioproducts with value added properties, opening up new possibilities for structural and functional versatility. The outstanding physical qualities, mechanical strength, biocompatibility and renewable source of production have led to the widespread application of NC in food packaging, biomedical science, mechanically reinforced polymer composites, paper and pulp, antibacterial coating material and environmental remediation and optoelectronics, among others [8,108]. Moreover, an intriguing aspect of NC is the presence of an abundance of functional groups on its surface, which allow for chemical changes and are further tunable in accordance with future application requirements. Numerous applications of nanocellulose can be facilitated by the intermolecular electrostatic attraction on nanocellulose surfaces.

#### 5.1. Biomedical Applications

Nanocellulose is regarded as one of the most distinctive and economical natural nanomaterial, with widescale biomedical applications within the health sector. It has remarkable uses, including the creation of scaffolds, applications for drug delivery, biosensors, the dressing of wounds, diagnostic tools, and the most cutting-edge methods of tissue and cell engineering [109]. Pharmaceutical industries frequently use nanocellulose due to it being renewable, non-hazardous, biocompatible and outstandingly durable. In a recent study, nanocellulose isolated from chili post-harvest biomass residue was blended with the extract from *Hemigraphis colorata* that displayed efficient antimicrobial activity by inhibiting the growth of *Bacillus cereus*, *E. coli*, *Pseudomonas* sp. and *Saccharomyces cerevisiae*, showing its potential to be used as a wound dressing material [110].

Nanocellulose has also been used in the medical industry for drug delivery to specific cells and soft tissue implants. Dehkhoda et al. extracted carboxylated nanocellulose from oat husk waste biomass using ammonium persulfate oxidation, and evaluated its in vitro indomethacin drug release [111]. The results exhibited a 67% drug release in 12 h, where the chemically modified nanocellulose significantly acted as controlled indomethacin

release carrier. Cellulose nanocrystals can be used to prepare natural hydrogel film that has the potential to be used as a drug carrier and controlled release material. Cellulosic materials, such as nanocellulose, can also be combined with the cyclodextrins in better drug delivery systems. Cyclodextrins are biologically compatible with cellulosic materials such as nanocellulose, and can therefore be covalently bond to the surface of cellulose to make the cavity of cyclodextrin accessible to guest molecules [112]. Nanocellulose is also regarded as a special scaffold material divided into three types, i.e., membrane scaffolds, bio-composite matrix and pellicle scaffolds [109]. Studies have also shown that nanocellulose in biomedical applications is nontoxic to cells, confirming its potential future applications in medical science.

### 5.2. Wastewater Treatment Using Nanocellulose

The development of treatment methods for wastewater management is of great interest on a global scale, to guarantee the availability of clean water. The release of heavy metals has negative effects on the ecosystem, economy and society. Due to their high aspect ratio and easy accessibility of numerous -OH groups for binding with dyes, heavy metals and other pollutants, nanocellulose excels in this specific application of waste water treatment [113]. Reports have suggested that various heavy metal ions can be extracted using nanocellulose-based material systems. As a result of the availability of numerous reactive groups and highly specific surface areas, the surface modification of nanocellulose improves its binding performance. Different techniques, such as sulfonation, amination and carboxylation are the techniques used to enhance the adsorption efficacy of nanocellulose for heavy metals in wastewater [114]. The effectiveness of rice straw fibers, cellulose fibers and nanocellulose fibers in removing metal ions was examined by Kardam et al. [115]. They conducted sorption experiments against Pb(II), Ni(II) and Cd(II) at a metal concentration of 25 mg/L (pH 6). They discovered that at a low dose, nanocellulose fibers outperformed cellulose and rice straw fibers in terms of removal effectiveness. Wang et al. also reported the use of cellulose nanofibers with PVAm grafting to remove Cr (IV) from water [116]. It has also been found that nanocellulose composites have applications in the bioremediation of organic xenobiotic compounds in complex environments [117].

### 5.3. Applications of Nanocellulose in the Food Industry

Nanocellulose is quite simple to use in the food industry due to its large aspect ratio, surface area, non-cytotoxic, rheological behavior and non-genotoxic properties [109]. In the food industry, nanocellulose finds a variety of applications, including as food stabilizers, thickeners, nutritional fibers, suspension stabilizers, flavor carriers and as a way to reduce calorie content. Additionally, it can be used to make soups, cookies, crisps, puddings, fillings, wafers, ice cream, crushes, etc. [118]. Nanocellulose is also considered a non-caloric stabilizing agent and a gelling agent in food applications due to its high viscosity even at lower concentrations [119]. When used as nanofillers in packaging film, the nanocellulose materials have a tremendous potential to sustainably enhance the tensile and barrier properties. Microfluidization was used by Bai et al. to create extremely stable Pickering emulsions with excellent properties from a variety of oils. Intriguingly, droplets with crystalline nanocellulose showed improved flocculation resistance under different conditions [120].

Nanocellulose can also be employed as a food additive in different food products. A study was carried out to assess the effect of CNC on the gelatinization, retrogradation characteristics and pasting of maize starch, waxy starch and sweet potato starch [121]. The results demonstrated that the addition of CNC improved the peak viscosity of sweet potato starch and maize starch, while reducing the setback values of sweet potato starch. In another study, Liu and Kong investigated the effect of NC in improving the intestinal digestion of milk and starch, besides the absorption of minerals. The results showed that CNC and CNF oxidized by TEMPO caused a significant reduction in glucose diffusion. Furthermore, the amount of fatty acids generated during the interval of milk fat digestion

was significantly reduced by CNF and TEMPO-mediated CNF. Additionally, the CNC reduced the rate of amino nitrogen diffusion during the breakdown of milk proteins [122]. CNCs are also good candidates for usage as a food additive that might replace shortening or partially hydrogenated oils with CNC, enhancing food safety and quality while reducing the detrimental effects of high-fat-based products on human well-being. In a work by Deloid et al., a 50% reduction in triglyceride hydrolysis was seen in the simulated gastrointestinal tract, which may be caused by fat droplets clinging to NC, thereby reducing the amount of fat which is accessible for lipase activity or preventing the solubility of fat digestion products [123].

The use of nanocellulose as a food stabilizer extends the shelf life and preserves the form of frozen dessert. Increasing the quantity of food stabilizers could improve the ability to maintain shape using stabilizers, but doing so would change the dessert's texture and taste. In order to determine how CNF would affect the composition and functionality of the frozen food, Velásquez-Cock et al. examined the addition of cellulose nanofiber to ice cream to check its impact on fat structure and relation to its adsorption. The addition of cellulose nanofiber improved the sensory qualities of reduced fat samples even after the specimens had been heat-shocked. Moreover, its addition had no effect on the hardness, rheology, or ice crystal development on the ice cream [124]. These studies suggest that NC can potentially be used as a food additive, packaging materials for the development of novel food materials.

#### 5.4. Nanocomposite Formation

In the past few decades, nanocellulose has drawn a lot of interest as a potential reinforcement for different polymeric materials or nanocomposites in a variety of cutting-edge applications. The nanocomposites derived from NC have gained much research interest to be applied in different fields owing to their unique properties, such as biodegradability, non-toxicity, renewability, highly specific surface area, high modulus and mechanical strength, and exciting optical properties and surface chemistry [125]. NC-based composites, in contrast to conventional cellulose-based composites, contain less well-dispersed nanocellulose, the reason being that it is not necessary to load the polymer with a lot of filler in order to get high mechanical properties [126]. Noguchi et al. prepared NC-reinforced polyethylene composite sheets using polyethylene and in situ surface-hydrophobized, TEMPO-oxidized cellulose nanofibrils using high shear forces [127]. In another study, cross-linked poly([2 (methacryloyloxy)ethyl]trimethylammonium chloride) ( $C_9H_{18}ClNO_2$ ) and bacterial nanocellulose were combined to create anti-fungal nanocomposites for the treatment of *Candida albicans* infections [128]. A variety of different nanomaterials can be impregnated into nanocellulose to create highly porous substrates.

#### 5.5. Nanocellulose in Printed Electronics

Paper is a desirable material for printed electronics because it is formed of cellulose, which is a recyclable, renewable, biodegradable and non-toxic substance. Nanocellulose obtained from cellulose has recently been shown to have enormous potential applications in flexible-printed electronics. Since NC has the ability to build self-standing thermally stable films, it has been used to create translucent and smooth substrates for printed electronics [129]. The performance of printed devices can be enhanced while staying environmentally benign by using nanocellulose to regulate the surface, speed up ink-drying and hold functional ingredients [130]. Through the inventive use of nanocellulose as the substrate and the transfer printing technique, Mao et al. established a type of ultrathin and reusable liquid metal circuit [131]. Likewise, cellulose nanofibers were applied as substrates for printed electronics by Penttila et al., where the findings indicated that the conductivity of printed lines on kaolin surface is greater than the conductivity on precipitated calcium carbonate surface [132]. Such materials prepared from nanocellulose might not function as well as their pure inorganic equivalents; however, the low cost and renewability of materials for products with short lifespans make them ecofriendly and compatible. It

is necessary to conduct more research on substrate development to enable back-to-back printing through the production of nanocellulose-based paper in printed electronics.

## 6. Agro-Industrial Food Waste for Nanocellulose Production

### 6.1. Agricultural Waste in Circular Economy

In the 21st century, population growth has raised the need for food and energy, which has resulted in a rapid depletion of natural resources over time. This increase in population and demand for resources has put pressure on the food processing industries, which produce large amounts of plant, animal and agricultural residue wastes, thereby accumulating a huge amount of waste throughout the year due to a lack of effective waste management methods [10]. The majority of food waste is produced during its production, harvesting, transportation, industrial processing and consumption [22]. According to estimates, one-third of all food produced worldwide is wasted or lost, with the fruit, vegetable and seafood industries playing a significant role in this. An estimated  $1.3\text{--}2.0 \times 10^9$  tons of waste (between 33% and 50% of the world's food production) is produced by the food industry annually, with fruit, vegetables and their by-products contributing to the largest amount of approximately  $0.5 \times 10^9$  tons per year. Around 100 Mt of food waste and by-products are produced annually solely in Europe, with the fruit and vegetable industry accounting for 14.8% of this total [22,133,134]. Moreover, residues from the agriculture industry (agro-industrial waste), which is also one of the types of LB, plays a significant role in waste generation. A total of 998 million tons of agricultural residues of all kinds are produced per year throughout the world, which includes rice/wheat straw, corn stover and a number of other economic crop leftovers [135]. This waste does not only exacerbate the food deficit, but also harms the environment when improperly disposed of, contaminating the land and water sources. Due to a number of obstacles, including the product's stability, pathogenicity, auto-oxidation potential, high water content and strong enzymatic activity, disposal of this waste incineration and landfilling is perilous. Waste products from the agriculture and food industries often contain useful substances that can be recovered and used to produce environmentally benign and viable products.

The concept of "circular economy", which advocates using organic waste from one industry as a source of raw materials for another for the production of value-added products, has gained significant interest in recent years. It is based on the implementation of the 5Rs principles of sustainable development, which are reduce, reuse, recycle, recovery and restore. These principles overcome the traditional practice of "take, make, use and throw" [4,22]. Therefore, following the practice of "circular economy", the agro-industrial waste, being rich in carbohydrates, proteins, lipids, organic acids and other necessary minerals, can be directed toward the production of sustainable products for its value addition. The exploitation of residual agricultural processing wastes is still in its infancy and is primarily defined by low value-added applications, such as direct combustion for heat production or use as a fertilizer/compost. Nevertheless, from economic aspects or environmental advantages outlook, these uses have not yet fully realized their potentials. Moreover, apart from the renowned potential of agricultural waste in producing biofuels, it can also be used to produce NC that possess promising applications in cosmetics, drugs, biomedicine and many other industries [18,136].

### 6.2. Production of Nanocellulose from Common Agro-Food Wastes

A number of commonly generated agro-industrial food wastes have been employed by different researchers to extract cellulose and nanocellulose, using a range of pretreatment technologies. Zheng et al. [137] produced NC from walnut shells (WSs) using 2,2,6,6-tetramethylpiperidine-1-oxyl radical (TEMPO;  $(\text{CH}_2)_3(\text{CMe}_2)_2\text{NO}$ ) oxidation and the sulfuric acid hydrolysis method. The cellulosic fraction was extracted using NaOH, followed by bleaching. WS biomass was pretreated using 2% NaOH at a solid-to-liquid loading of 10 g/100 mL, and pretreatment was carried out for 4 h at 100 °C under constant stirring. TEM analysis revealed that the NC produced via TEMPO oxidation was irregular-block in

shape, whereas sulfuric acid NC was 55–82 nm in length and 49–81 nm in width [137]. In another study, the production of NC from pineapple leaves was demonstrated. Pineapple leaf biomass was first pretreated using 4% NaOH at 80 °C for 2 h, followed by bleaching. The CNC from alkali-pretreated pineapple leaves was extracted using 64% (*w/w*) sulfuric acid (10 mL) at 45 °C for 60 min. The CNC produced was rod-like in structure, about 100–150 nm in diameter and 3–5 nm in length, and was successfully utilized as reinforcing fillers in natural rubber (NR) composites [138]. CNC production was also carried out from Vietnamese agricultural wastes (*Nypa Fruticans* trunk, coconut husk fiber, and rice husk). The biomasses dispersed in distilled water were heated at 100 °C for 2 h for the removal of impurities, followed by oven-drying. The biomasses were then pretreated with 90% HCOOH at 100 °C for 2 h under a magnetic agitator in the circulation system. The hemicellulose content was removed by dispersing biomasses in a mixed solvent of formic acid, hydrogen peroxide and distilled water (90:4:6% *w/w*, respectively) at 80 °C for 2 h, followed by filtration and washing using formic acid (80%) and distilled water. The CNC was produced using 64% sulfuric acid solution at 45 °C for 30 min with a strong agitation in the circulation system. The length and width of the CNC obtained was 200–500 nm and 10–15 nm, respectively [139]. Table 2 also lists some other agro-industrial food wastes which have been reported for the production of NC. For the sustainable production of NC from LB, it is necessary for a waste biomass to be abundantly present, easily transported from the fields of the industry for processing, and it must be rich in cellulose content that is considered a precursor for NC.

**Table 2.** Production of different NCs from various agro-industrial residues.

Agro-Industrial Food Waste	Cellulose Content	Pretreatment Condition	Method of NC Production	Type of NC	Dimensions	Reference
Tea stalk	35.01%	H <sub>2</sub> O <sub>2</sub> and H-Ac solution (1:1): 60 °C, 24 h	Acid hydrolysis using 60%–65% H <sub>2</sub> SO <sub>4</sub>	NCC	Width = 4–8-nm	[140]
Rice straw	36.5 ± 2.1%	Mutistep purification; 6% NaOH at 60 °C for 2 h; 30% of H <sub>2</sub> O <sub>2</sub> at 90 °C for 5 h	Ammonium persulphate method	CNF	Diameter = 14 ± 7.0 nm	[141]
Rice straw	36.5 ± 2.1%	Mutistep purification; 6% NaOH at 60 °C for 2 h; 30% of H <sub>2</sub> O <sub>2</sub> at 90 °C for 5 h	Ammonium persulphate method	CNC	Diameter = 19 ± 5 nm; Length = 118 ± 52 nm	[141]
Apple pomace	32.48 ± 0.33%	Heated at 90 °C in distilled water for 30 min; 1 M HCl at 85 °C stirred for 30 min; repeated twice with 0.5 M HCl	Acid hydrolysis using 45% H <sub>2</sub> SO <sub>4</sub>	CNC	Diameter = 7.9 ± 1.25 nm; Length = 28 ± 2.03 nm	[142]
Coffee husk	35.4 ± 0.9%	4 wt% NaOH solution, at reflux temperature for 3 h, under continuous stirring	Acid hydrolysis using 64% H <sub>2</sub> SO <sub>4</sub>	CNC	Diameter = 20 ± 4 nm; Length = 310 ± 160 nm	[143]
<i>Salicornia ramosissima</i> waste	17.6 ± 0.6%	15% NaOH (1:15 <i>w/v</i> ) and 30% H <sub>2</sub> O <sub>2</sub> (100:1.2 <i>v/v</i> ) under constant stirring at 50 °C for 3 h	Acid hydrolysis using 2% H <sub>2</sub> SO <sub>4</sub>	CNF	Diameter = 17.9 ± 2.8 nm; Length = 928.6 ± 38.5 nm	[144]
Orange bagasse ( <i>in natura</i> )	11.85 ± 2.73%	NaOH (1.6%, 2%, 3% and 4%, <i>w/v</i> ), at 120 °C, 1 atm pressure for 20 min	Ultrasound treatment	CNF	Diameter = 18.4 nm ± 6.0 nm	[145]
Industrial orange bagasse	21.04 ± 7.43%	NaOH (1.6%, 2%, 3% and 4%, <i>w/v</i> ), at 120 °C, 1 atm pressure for 20 min	Ultrasound treatment	CNF	Diameter = 20.5 nm ± 7.0 nm	[145]
Pineapple Leaf Fibers	62.5%	25% NaOH at 150 °C, 9 bar for 60 min	High-shear homogenization and ultrasonication	CNF	Diameter = 68 nm; Length = 88–1100 nm	[146]
Corn cobs	45.01 ± 0.9%	Microwave-assisted delignification: 80 W power, 12.86% Sodium chlorite for 16 min; Ultrasound-assisted alkali extraction: 14% NaOH at 41 °C for 46 min	Acid hydrolysis using 60% H <sub>2</sub> SO <sub>4</sub> and ultrasonication	CNC	Diameter = 131.4 nm	[65]
Garlic straw residues	41%	2 wt% NaOH, 12 h	Acid hydrolysis using 65% H <sub>2</sub> SO <sub>4</sub>	CNC	Diameter = 6 nm; Length = 480 nm	[147]
Grape pomace	19.30 ± 0.67%	Acid pretreatment: 2% H <sub>2</sub> SO <sub>4</sub> at 90 °C for 5 h under constant stirring; Alkaline pretreatment: 5% NaOH at room temperature for 10 h and continued at 90 °C for 5 h	Acid hydrolysis using 64–65% H <sub>2</sub> SO <sub>4</sub>	CNC	Diameter = 7 nm; Length = 323 nm	[148]
Wheat straw	44.81 ± 0.67%	Steam explosion: 100 °C, 3.0 MPa pressure for 2 min; Acid pretreatment: 1 M HCl at 80 °C for 2 h under stirring; Microwave-assisted alkali hydrolysis: 2% NaOH, 40 °C, 1200 W for 20 min	Microfluidization treatment	CNF	Diameter = 5.42 nm; Width = 10–40 nm	[149]
Sugarcane bagasse	42 ± 2.5%	10% ( <i>w/w</i> ) NaOH for 1 h at 80 °C, under mechanical stirring	Enzymatic hydrolysis using Cellic CTec3 (Novozymes, Bagsvaerd, Denmark)	CNC	Diameter = 12.2 ± 5.6 nm; Length = 506.1 ± 123.7 nm	[150]

NC = nanocellulose; NCC = nanocrystalline cellulose; CNC = cellulose nanocrystals; CNF = cellulose nanofibril.

## 7. Conclusions and Future Perspectives

The current review reports various methods for the extraction of nanocellulose from waste lignocellulosic biomass, and different possibilities for its application in a range of commercial industries. Green bio-based renewable raw materials and more ecologically friendly and sustainable resources are currently one of the major sustainable development goals set forth by the United Nations. Since cellulose is the most prevalent source on earth, it has undergone extensive research by scientists from all over the globe. Nanocellulose has attracted a lot of interest in the recent years because of its biocompatibility, renewability, bioavailability and other noteworthy characteristics. The removal of all non-cellulosic materials requires the biomass to be pretreated because lignin, hemicelluloses and other materials firmly aggregate in the cell walls of waste biomass. Pretreatment of lignocellulosic biomass is quite necessary for the extraction of cellulose, removal of lignin and other impurities, which enhance the quality of crystalline nanocellulose and nanofibers. Their structural and physicochemical characteristics are significantly influenced by the method used to separate nanocellulose.

Compared to traditional extraction techniques, green cellulose extraction technologies from agro-industrial food waste have a number of benefits. The advancements in cellulose recovery employing ultrasonic and microwaves can lower the treatment time and the quantity of the energy used, in line with ecological and sustainable goals. The use of mild acid could cut down on the water needed to get the material pH level to neutral. DESs have been suggested as safe compounds for cellulose extraction from agro-food waste. The ongoing studies in this area demonstrate how reliable and repeatable procedures are necessary to guarantee the adoption of nanocellulose within the industry. Cellulose nanomaterials have received an extensive interest as biomaterials, with a variety of applications in different food industries. However, it is essential to evaluate the material safety and any negative impacts on human beings and the environment before developing nanoscale materials and bringing them into the market.

The development of nanocellulose from waste biomass has opened up new avenues for scientific study, as well as engineers working in the area of nanoscience and nanotechnology. One of the main problems that restrict nanocellulose applications is the low thermal stability of cellulose materials. However, different methods are being adopted to improve the thermal stability of nanocellulose, including the removal of toxic acid groups and neutralization by alkaline solutions. The structural and functional characteristics of cellulosic derivatives are being investigated in numerous other research studies, such as alternatives to conventional packaging materials and metal ion detection, besides their major applications in drug delivery, tissue engineering and wastewater treatment. Undoubtedly, there are obstacles to be surmounted in order to produce innovative materials for sustainable applications. Therefore, more effective research outputs are required for the wider application range of nanocellulose-derived materials for sustainable development.

**Author Contributions:** Conceptualization, P.N. and C.-H.K.; Validation, V.S. and P.N.; Formal Analysis, V.S. and P.N.; Investigation, V.S.; Writing—Original Draft Preparation, P.N.; Writing—Review and Editing, H.-M.D.W. and C.-H.K.; Resources, M.-L.T. and S.-L.H.; Supervision, C.-D.D. and C.-H.K.; Funding Acquisition, C.-H.K. All authors have read and agreed to the published version of the manuscript.

**Funding:** This work was supported by research funding grants provided by the National Science and Technology Council of Taiwan (MOST 111-2221-E-992-005-MY3).

**Institutional Review Board Statement:** Not applicable.

**Informed Consent Statement:** Not applicable.

**Data Availability Statement:** Not applicable.

**Acknowledgments:** Author P.N. thankfully acknowledges the National Kaohsiung University of Science and Technology, Kaohsiung City, Taiwan, for providing the post-doctoral fellowship.

**Conflicts of Interest:** The authors declare no conflict of interest.

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