Extraction of cellulose nanocrystals from agricultural by-products: a review

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ABSTRACT
In the latest studies, the reuse, recycle, and recovery of various biomass wastes for high value-added bioproducts have received significant attention to reduce the environmental impact of fossil-based polymers. This review briefly describes various agricultural wastes used to extract cellulose nanocrystals (CNCs) and the pros and cons of common extraction methods. CNCs have been employed in high-performance materials due to their attractive properties such as lightweight, excellent mechanical strength, biocompatibility, biodegradability, renewability, high surface area, and tunable surface chemistry. Subsequent chemical modifications of CNCs allow it to serve in a wide range of applications: biocomposites, biomedical industry, food industry, cosmetics, wastewater treatments, and electronics industry. The challenges of CNCs production, the deficit in physicochemical properties and applications, and prospects of CNCs are discussed in detail in this review.

ARTICLE HISTORY
Received 30 May 2022
Accepted 31 August 2022

KEYWORDS
Agricultural wastes; cellulose; cellulose nanocrystals; extraction; surface functionalization

1. Introduction
As petroleum-based resources stay in the environment for longer period without degradation, recent studies focused on searching more environmentally friendly alternatives (1). Besides the environmental impact, crude oil is also depleted due to its non-renewability. Thus, a bio-based raw material source shows a large potential to be renewable for the production of high added-value materials or biopolymers if adequate care is taken during their exploitation. The biomass sourced from the structural portion of plants that includes agricultural and forestry residues are the most abundant organic materials in nature that comprise about 40–50% cellulose and 20–30% hemicellulose, with lesser amounts of lignin and other compounds such as sugars, oils, and minerals (2, 3). Cellulose is among the most promising sustainable biomaterials due to its intrinsic properties such as abundance and renewability if care is taken during its harvest (4, 5). Cellulose is a polysaccharide organic compound consisting of linear chain of β(1→4) linked D-glucose units and abundant hydroxyl groups on its surface which has a formula of (C₆H₁₀O₅)ₙ and structure shown in Figure 1.

Nanocellulose, an eco-friendly and promising natural material, is isolated from cellulosic sources through chemical, mechanical, or biological extraction methods. Based on material sources, morphologies, and preparation methods, nanocelluloses can be divided into three main categories: cellulose nanocrystal (CNC), cellulose nanofibril (CNF), and bacterial nanocellulose (BNC) (6). CNC and CNF are isolated by partial disruption of cellulosic structures through chemical and/or mechanical
treatments. Whereas, BNC is synthesized by certain gram-negative bacteria via polymerization of glucose. Compared to the ordinary cellulose, CNCs have excellent mechanical strength, large surface area, and an ideal length-diameter ratio (7). At present, most commercially available CNCs are mainly produced from high-quality raw materials such as cotton, wood pulp, and microcrystalline cellulose. However, recent studies by many researchers confirmed that biomass wastes are also considered as ideal materials for the extraction of high value-added products such as CNCs (8–10). Among several biomass wastes, agricultural wastes show large potentiality for the production of CNCs by using different extraction methods. Agricultural wastes of about 998 million tons were produced from various crops and lignocellulosic fibers throughout the world annually (11). Several agricultural wastes such as cocoa pod husk (12), raw rice husk/straw (13, 14), date pits (15), enset fibers (16), khat waste (17), coconut husk (18), jute fibers (19), ramie fibers (20), corn cobs/husk (21), kenaf bast fibers (22), wheat straw (23), pea hull (24), pineapple leaves (25–27), raw apple stem (28), and many more have been utilized for the extraction of CNC. Figure 2 depicts some of the most common agricultural wastes used for the extraction of CNCs.

Before the actual extraction of CNCs, a native cellulose is obtained by removing the extra components from these wastes through pretreatment processes. The most common pretreatment methods are alkaline treatment and bleaching process (29). During the alkaline treatment, non-cellulosic components such as hemicelluloses and pectins are fully solubilized whereas lignin is partially depolymerized. In the bleaching process, chemicals such as calcium hypochlorite (CaClO₂), sodium chloride (NaClO₂), or hydrogen peroxide (H₂O₂) are used to completely remove lignin and whitening of cellulosic fibers. Accordingly, the pretreatment processes make the cellulosic part more accessible for further treatment during the extraction of the nanocelluloses. CNCs extracted from these wastes have valuable properties such as biodegradability, biocompatibility, low density, excellent mechanical properties, chemical inertness, renewability, low thermal expansion coefficient, stable structure, high specific area, and low cost as well (7, 30).

Even though many agricultural by-product’s ability to produce nanocellulose were described by several researchers, numerous wastes are not investigated yet. As an example, agricultural wastes such as teff straw, shimbira stem and hull, guayo straw and hull, enset fibers, and stem, and coffee husk are among the by-products generated in Ethiopia but dumped into the environment without changing into any value-added products. This indicates that there is a wide opportunity to undertake research in this area to obtain a simple, cost-effective, and batch process of CNC production. Thus, this study was designed to review the potentiality of converting agricultural wastes to CNCs, their potential application areas, analyzing the current challenges, and suggesting future prospects in detail.

2. Extraction of CNCs

The extraction of CNC from plant fibers is a process of forming a highly crystalline cellulose structure by removing the amorphous parts of the cellulose. Based on the material sources and extraction methods, nanocelluloses can be divided into three major categories: CNCs, cellulose nanofibers (CNFs), and BNCs. The overall characteristics of the extracted nanocelluloses such as particle size, crystallinity, and morphology are varying from one another due to the difference in the cellulose origin, extraction and processing conditions, and possible pre- or post-treatments. The most common method of obtaining the CNC from different biomass wastes by decreasing the size of natural cellulose is shown in Figure 3. These include the mechanical process, chemical hydrolysis, biological hydrolysis, and combined methods (31).

In the mechanical process of CNCs extraction, the cellulosic sources will be subjected to high shear forces such as ultrasonication, micro-fluidization, cryo-crushing, high-pressure homogenization, grinding, and ball milling (32). This method alone was not widely applied due to its great energy consumption and the morphology of cellulose is also affected by mechanical operations, which in turn resulted in a lower degree of crystallinity and purity of extracted CNC (33, 34). Another method is chemical hydrolysis: the crystalline parts of the cellulose remain intact whereas the amorphous fraction is dissolved in this process. It is among the low-cost and convenient methods for preparing the CNC. However, it has disadvantages such as thermal instability due to the presence of sulfone groups on the extracted CNC surface, requiring a high amount of water and special reactors to separate acid.
or alkali from CNCs and neutralization of wasted solutions that are harmful to the environment (6).

The third method, called biological hydrolysis, also known as BNC, is mostly fermented from certain gram-negative non-pathogenic bacteria such as Acetobacters species via polymerization of glucose and crystallization. This process consumes very low energy, eliminates the use of harmful chemicals, and results in a highly

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**Figure 2.** Some of the common agricultural wastes used for CNC extraction: (a) coconut husk, (b) cocoa pod husk, (c) corn cob, (d) ramie fiber, (e) enset fiber, (f) kenaf fiber, (g) rice husk, (h) wheat straw, (i) jute fibers, (j) pineapple waste, (k) date pits, and (l) pea hull.

**Figure 3.** Schematic diagram of common CNCs extraction methods from agricultural wastes: adapted from (31).
effective and biocompatible material which fulfills the requirements of green and sustainable development (35, 36). However, BNC is highly sensitive process for synthesis parameters such as culture medium, bacteria strain, inoculum concentration, temperature, and fermentation time (37). The commonly used culture medium called Hestrin-Schramm (HS) (38) makes a synthesis of BNC costly process as it represents up to 30% of the total production costs (39). To overcome the problem of high culture medium costs, several researchers used agro-industrial wastes as an alternative source of carbon with low-cost fermentation media to replace the D-glucose in HS medium (40–42). The BNC possesses diverse physicochemical properties and morphologies based on the types of bacteria used under a variety of culturing conditions and intended application areas. The use of fine and expensive culture medium constituents, low efficiency, strict requirements of timing, and reaction conditions mainly limits the large-scale productivity and competitiveness of the BNC (43, 44). The fourth type of extracting CNC is known as a combined method in which the chemical or biological methods are integrated with physical methods to produce the nanocelluloses for various purposes (45, 46). In this process, the alkaline acid or mild enzymes pretreatment will be employed to solubilize the hemicellulose, lignin, and pectin before the implementation of the mechanical process for the extraction of CNF.

The overall characteristics of CNC extracted from different agricultural wastes by different researchers are compared in Table 1.

### 3. Surface modification of CNCs

The extracted CNCs always have a large number of hydroxyl groups that oblige them to attain a hydrophilic nature. The presence of these functional groups is very helpful when it is used in aqueous media such as reinforcing hydrophilic polymers. However, the direct use of these hydrophilic CNCs in hydrophobic matrices challenges its uniform dispersion which results in the lack of interfacial interaction, agglomerate formation, and phase separation. Thus, uniform dispersion of CNC is very critical to obtain materials with improved mechanical properties.

To improve the applicability of CNCs in a variety of fields, physical, and/or chemical surface modification is necessary after the extraction process. These include processes such as oxidation, chemical grafting,
esterification, silylation, sulfonation, and acetylation as shown in Figure 4. These processes do not harm the crystalline nature of the nanocellulose when executed under controlled conditions to form a hydrophilic–hydrophobic balance in a selected matrix.

3.1. Oxidation

This method is used to modify the hydrophilic surface of CNC to facilitate better dispersibility in aqueous media. The most commonly used oxidation is TEMPO-mediated oxidation. The presence of TEMPO typically converts the hydroxyl groups present on the C-6 of the glucose unit into negatively charged carboxylate groups. The introduction of dense carboxylate groups on the CNCs surfaces due to the TEMPO oxidation helps to obtain uniform and transparent aqueous suspensions of the nanocelluloses without changing their original morphology (27, 63–66). CNCs extracted from several agricultural wastes such as corn husks, banana stems, blueberry residues, oil palm, and pineapple leaves are subjected to TEMPO oxidation and show a width between 1.6 and 10 nm and a few microns in length. Ammonium persulfate (APS) is also used as an oxidant, one-step hydrolysis of cellulosic sources to produce CNCs with dense carboxyl groups surface (67). The APS and TEMPO-mediated oxidation of the CNCs reduces the amount of corrosive chemicals required for the treatment and energy involved in the mechanical extraction process. This results in the reduction of the environmental impact and the cost of production of the CNCs.

3.2. Sulfonation

Sulfuric acid is used to catalyze the CNCs hydrolysis process to replace the hydroxyl groups of nanocellulose with sulfate groups. The replacement of hydroxyl groups with sulfate ester groups on the surface of CNCs restrains the hydrogen bond formation between cellulose molecules and improves its dispersion in aqueous media by forming a stable colloidal suspension without the evolution of CNCs aggregates (68). However, forming dense sulfate groups on the surface of CNCs by the sulfonation method is challenging and it may require further modification like TEMPO oxidation after the sulfonation process. The presence of dense sulfate groups on the nanocellulose surface helps to produce uniform dispersibility of CNCs in an aqueous medium, which makes them suitable for a wide range of applications.

3.3. Acetylation

This is among the well-known methods to reduce the hydrophilicity of nanocellulose by introducing nonpolar ester groups mostly by using acetic anhydride due to its simplicity and cost-effectiveness (69). The reduction in hydrogen bonds between cellulose molecules were observed after the acetylation process, which in turn
improves the dispersion of nanocelluloses in nonpolar matrices (70). In the presence of a catalyst such as sulfuric or perchloric acid, the acetylation reaction further accesses the hydroxyl groups buried inside the cellulose crystals to substitute them with acetyl groups. Thus, acetylation causes an increase in hydrophobic character, thermal stability, and better control of CNCs size. However, the acetylation undertaken in acid hydrolysis leads to a reduction in crystallinity and changes in the original morphology of CNCs (71). To improve those negative effects, simultaneous hydrolyzing of the cellulose amorphous parts and acetylation of the hydroxyl groups by using vinyl acetate in a single step are developed by several researchers. Despite the excessive acetic acid waste generated, acetylation is among a good example of a sustainable CNCs surface modification since it can be undertaken without the need of any additional catalyst or activation step.

3.4. Polymer grafting

A wide variety of functional polymers can be grafted to the abundant hydroxyl groups on the CNCs surface by forming covalent bonds in two approaches: ‘grafting to’ and ‘grafting from’ as shown in Figure 5 to control its dispersibility, interfacial and self-assembling properties (72). In the grafting to approach, a pre-functionalized polymer is covalently bonded onto the CNC’s surface where there is low grafting density due to the high steric hindrance. Whereas, the grafting from can be initiated by ring-opening polymerization where the hydroxyl group on the surface of CNCs is used as in-situ site for initiating polymerization. Polymers such as polyactic acid, polyethyleneimine, polycaprolactone, maleated polypropylene, and polyurethanes are commonly used in either grafting from or grafting to approaches with CNCs (73, 74).

The grafted CNC presents good dispersion in polymeric matrices due to the reduction in the surface energy with in turn results in a better thermal stability of the CNCs. Several researchers proposed the synthesis of grafted CNC via free radical strategy and ozonolysis reaction steps that are consistent with principles of green chemistry (75). The initiator-free environmentally benign gamma radiation is employed to graft poly-acrylic acid onto corn husk-based materials to develop promising, cheap, efficient, and reusable dye adsorbents (76).

3.5. Silylation

The reaction between silane coupling agent and hydroxyl groups is undertaken to introduce the hydrophobic alkyl groups on the nanocelluloses surface. The Si–O–C bond formed during the de-polymerization reaction help to achieve better interfacial adhesion between the CNCs and polymeric matrix (62). Figure 6 shows the condensation reaction between hydroxyl groups of the CNCs and isocyanate groups of 3-isocyanatopropyltriethoxysilane (IPTS). Here, the IPTS was grafted on the surface of the CNCs in dispersed anhydrous N,N-dimethylformamide (DMF) by sonication at 80 °C in the sequentially added tin(II) ethylhexanoate (Sn(Oct)2).

The silylated cellulose nanocrystals (SCNCs) showed much better dispersion, good compatibility, improved interfacial interaction, and a stronger reinforcing effect within the polymeric matrix than pure CNC (77). A direct, simple, and straightforward method to functionalize the surface of CNCs with 3-aminopropyltriethoxysilane (APTES), without using hazardous solvents was undertaken by Khanjanzadeh et al. (78). The CNC dimensions and crystalline structure did not show significant changes as a result of the modification. Compared to non-modified CNC, silylated CNC exhibited good thermal stability and a greater amount of residual char at about 500°C.

3.6. Surfactant’s adsorption

Surface-active species of nonpolar media like surfactants, polyelectrolytes, or counter ion salts are adsorbed
to alter the surface chemistry of CNCs via non-covalent interactions such as electrostatic attraction, hydrogen bonds, or van der Waals forces. A surfactant, acid phosphate ester of ethoxylated nonylphenol, modified CNCs favored the dispersion of s-CNC in the PLA matrix and maintained the high strength and optical transparency for food packaging applications (79). To study the capability of CNC as a drug carrier, the adsorption of tetracycline onto CNC surface was conducted isothermally at 30°C under the static condition at varied pH (80). The in vitro release of tetracycline drug shows the highest release of tetracycline 82.21% at pH 7.2 and the lowest 25.1% at pH 2.1. This implies that CNC can be used as drug carrier without harmful chemical excipients that comply with health safety, biocompatibility, and biodegradability. Controlling the concentration of surfactant is vital as low concentration leads to ineffective CNCs dispersibility in organic solvents, whereas self-aggregation of nanocelluloses is induced in an excess amount during the mixing process. The covering of nanocelluloses with surfactants resulted in more hydrophobicity, lower surface tension, higher dispersibility, improved compatibility, synergistic stabilization, gel formation, and the highest oxidative stability (81).

4. Applications of CNCs

Various agricultural wastes have been utilized as raw material sources to extract cost-effective and high value-added products, CNCs. The development of CNCs from these wastes plays a critical role in economic, environmental, and social sustainability. CNCs have distinguished properties such as excellent mechanical properties, nontoxicity, biocompatibility, lightness, high water absorption and retention ability, high surface area, low thermal expansion coefficient, and renewability (2, 7, 30). Physical and/or chemical surface functionalization of CNCs tuned it to have a variety of functional groups with different properties. This enables them to be utilized in diverse applications such as composites, biomedical and pharmacy, packaging, food, cosmetics, optoelectronic devices, and many other potential applications. Figure 7 depicts some of the applications of nanocelluloses.

4.1. Nanocomposites

CNC is a green nanomaterial that can be utilized for nanocomposite formulations due to its excellent mechanical performance combined with its abundance, low density, nontoxicity, biodegradability, and sustainability. The introduction of CNC into biopolymers improved the performance characteristics and further sped up the biodegradation rate of nanocomposites (79, 82). Alana et al. showed that the incorporation of nanocelluloses extracted from agricultural and industrial waste in polylactic acid (PLA) improves the tensile strength from 63 to 78 MPa for neat biopolymers and nanocomposites, respectively (83). This enhancement is mainly related to the good dispersion of the CNCs and their interactions with the functional groups of PLA and also the stress transferability between the matrix and fillers. The incorporation of CNC in PVA significantly improved tensile strength of 140.2% when only 9 wt % CNC was incorporated without affecting the transparency or homogeneity of the polymeric matrix (53). Similarly, the incorporation of CNC in PVA/Chitosan retained the transparency and enhanced the tensile strength of the nanobiocomposite film with more than 21% (84). Furthermore, the prepared nanobiocomposite film exhibited good antimicrobial activity against foodborne pathogenic bacteria and postharvest pathogenic fungi. Another study reveals that the addition of 5 wt% CNC in maleic anhydride grafted PP showed optimum tensile strength and modulus up to 25.53% and also increase in stiffness of PP nanocomposites (74). Even though certain study reports that the light transmittance values were decreased due to the incorporation of CNC, but the mechanical properties were enhanced in all cases (85, 86). On the other hand,
functionalized CNCs have been utilized to produce nanocomposites for a variety of advanced applications such as nanomedicine, opto-electronics, environmental remediation, water treatment, catalysis, and many more applications (87).

4.2. Foodstuffs and packaging

The wastes generated after using conventional food packaging materials such as polyethylene or copolymer-based polymers have adverse environmental effects as it stays for long periods without degradation. Thus, replacing these hazardous packaging materials with biodegradable and renewable components not only sustains environmental safety but also helps to prevent food contamination by incorporating antimicrobial nanoparticles at their active sites (84, 88). The weak bonds and covalent intermolecular linkages formed between the nanocelluloses and polymeric matrix provide more flexibility as well as good strength properties with a defined polymorphic structure that permits thermoforming process of the packaging materials (89). The packaging made up of TEMPO-oxidized nanocellulose hydrogel has been used as a carrier for PH-responsive dyes to analyze the freshness of the chicken sample based on the color change (90). As the microorganisms level and spoilage of chicken increase, the nanocellulose hydrogel optical color changed from green to red, which shows that the acceptability limit for human consumption is passed. This indicates that the freshness of food can be quickly analyzed by utilizing nanocelluloses as an intelligent packaging material (91). On the other hand, CNCs have been added to food products to avoid food deterioration and extend the shelf life (92, 93), detect hazardous residues, improve the tensile strength and reduce moisture content and oxygen permeability of food packaging films (94).

4.3. Biomedicine and pharmacy

CNC exhibits excellent properties such as superior biocompatibility, inertness, nature, high mechanical strength, biodegradability, high surface area, and easy to surface modification which makes them an ideal material to be used for biomedical and pharmacy applications. The CNC-based polymer composites have large potential for the delivery of active molecules such as drugs, proteins, genes/plasmids, and tissue engineering. CNC is utilized in controlled and targeted drug delivery systems such as hydrogels, aerogels, nanoparticles, cryogels, and membranes due to its biocompatibility, high surface-area-to-volume ratio, low weight, efficient cellular binding, and

![Image of CNC applications](image_url)
uptake capacity (2, 95). Nanocellulose modified with antimicrobials and bioactive has been used in the delivery of various drugs for wound healing and hemostasis. The presence of CNCs in chitosan and/or polycaprolactone (PCL) nanocomposites allows sustained release of tetracycline drugs than pure chitosan and PCL due to its restrictions on the release of drugs (80, 96).

CNCs extracted from softwood and functionalized with acetyl trimethylammonium bromide were used for the controlled release of hydrophobic anticancer drugs such as docetaxel, paclitaxel, and etoposide where the drug was sustained effectively and killed the KU-7 cells for 2 days (97). Controled drug delivery has an advantage of releasing the drug that enhanced therapeutic effects with minimum side effects. The CNCs extracted from rice husks show antioxidant and antiproliferative to several phytochemical bioactivities such as p-hydroxybenzoic acid, caffeic acid, p-coumaric acid, ferulic acid, ad flavonoids (98). The human fibroblast skin cell incorporated on the surface of the polyvinyl alcohol (PVA)/CNC biocomposite didn’t show any adverse effects even after 24h of incubation (99, 100). This implies that the fabricated biocomposites had potential for use as biomaterial for tissue engineering where cells are properly adhered on its surface and live as healthy as in nature.

4.4. Electronics

CNCs have been recognized to achieve several low-cost smart materials due to their excellent properties such as high mechanical strength, high surface area to volume ratio, porosity, lightweight, thermal stability, eco-friendly, hydrogen-bonding capacity, and easy surface modification. CNC’s capabilities to be integrated into multiple components of electroactive materials allow it to form conductive and flexible electronic materials. As an example, many researchers have developed CNCs-derived carbon materials to produce highly electrically conductive materials that can be used as electron collectors in supercapacitors, sustainable energy storage, and sensor devices (101–104). The light weightiness, porosity, and strongness of nanocelluloses help to have a good ion and electron transferability by accommodating a high mass of conductive materials in CNC-based supercapacitors. In addition, CNCs showed large potential in the electronics field such as versatile electronics, sensitive hydrogels antireflective coatings, soft actuators, optical filters, and many more applications (2).

4.5. Wastewater treatment

The by-products released by human activities lead to the contamination of freshwater with various types of organic/inorganic compounds and microorganisms. These pollutants affect the water quality and result in harming human well-being, affecting aqueous habitats, disturbing biodiversity, and degrading the environment. Thus, many wastewater filtering techniques and membrane materials made up of CNCs have been developed by different researchers to mitigate the effects of these pollutants (6, 7). CNC is used as an efficient material for wastewater treatment due to its intrinsic features such as high surface area, surface charge, aspect ratio, mechanical strength, and stability in water. CNCs have been commonly used as green treatment strategies by adsorbing hazardous metal ions such as Ag+, Cd2+, Cu2+, Hg2+, Ni2+, Pd2+, and U6+ from contaminated wastewater (105–108). The hazardous metal ions sorption efficiency of nanocelluloses were derived from its high surface area and the existence of high hydroxyl (-OH) and modified surface such as carboxyl (-COO) groups that will have specific site interactions with specific metal ions. The higher the number of negative functional groups, the higher adsorption of hazardous metal ions. In addition to hazardous metal ions, modified CNCs based materials have been utilized for treating wastewater contaminated by hazardous organic compounds such as dyes (109), drugs (110), fertilizers (111), and pathogenic microorganisms (84) those are highly toxic to the environment.

4.6. Cosmetics

CNCs have been vastly used in skincare products as a carrier for specific ingredients or structuring agents in formulations due to their biocompatibility, surface functionality, dispersion stability, purity, high viscosity and shear thinning capacity, and water-holding capacity. CNCs were utilized as formulation of moisturizers, modifiers, additives, and nanofillers to synthesize bioactive compound carriers, UV-blockers, or skin sensors (112). Hemp CNCs grafted to polylactic acid (Hemp/CNCs-g-PLA) developed for skin protection resulted in an excellent performance in terms of biological compatibility, non-penetration into the skin, and water resistance (113). The Hemp/CNCs-g-PLA liquid-based foundation provides advantages such as avoiding discoloration, skin roughness, age spots, and easy-wiping characteristics that help to keep the skin from the damage aroused by excessive cleansing. Typically, BNCs show excellent properties in cosmetic applications as it highly supports skin active substances are accustomed as an emulsion stabilizer, and enzyme immobilizer that aids to treat skin dehydration, wrinkles, photoaging, and dark spots.
4.7. Paintings and coatings

CNCs have been employed in the production of paints as a coating agent to enhance the durability of paintings. CNCs introduced in painting demonstrated an improvement in color fastness to light, better smoothness of the paint, higher high shear tolerance, and good mechanical properties (114). Nanocelluloses also became a prominent material tailored to the surface of food as a microlayer film of edible coatings to preserve food such as perishable fruits and vegetables without quality decay during storage. CNCs incorporated in the polymeric matrix to form edible coatings improve the mechanical and barrier properties, stabilize and control the active agents release of edible coatings, which in turn resulted in the extension of the shelf life without deteriorating the quality of food (115). CNC coated on electrospun polyamide 6 (PA6) film resulted in lower porosity, higher surface roughness, and improved mechanical strength (21). The Young’s modulus of CNC/PA6 film has been dramatically increased by 250% which implies that the film can be used for infiltration and packaging applications. In addition to reducing the adverse gases for food packaging, the coated film added crucial values such as high transparency, thermal recyclability, and hostile to bacterial adhesion.

5. Challenges and prospects of CNCs

CNCs have been utilized to design several functional materials with high value-added and high performance due to their intrinsic properties such as low particle size, lightweight, high specific surface area, biodegradability, biocompatibility, nontoxic, high strength, easy surface modification, low cost, and environmentally friendly. These factors make CNC an important choice in various fields of biodegradable composites, biomedicine and pharmacy (scaffolds, drug carriers, and gene and protein delivery), food packaging, wastewater treatment, cosmetics, and electronic materials (2, 114, 116, 117). However, some challenges need to be overcome yet to sustain and promote CNCs in industrial applications and utilization: technical challenges and cost issues.

Currently, the cost of CNC is much higher than that of mineral nanofillers due to the high extraction costs and limited production capacity. The high cost of CNC is resulted from a series of actions like biomass waste collection and storage, use of expensive chemicals and enzymes; large energy consumption during the production processes (31). Even though the laboratory-scale costs of CNCs extracted from agricultural wastes are higher (~54 $/kg) than the costs of mineral nanofillers such as graphite and clay (~2–5 $/kg), it is much lower than that of pulp-based CNCs (118, 119). The current production costs of CNCs from agricultural wastes were only about 10% of pulp-based CNCs (~575 $/kg). Optimizing the process steps and obtaining the commercial-scale production of CNCs from agricultural wastes with excellent properties and uniform size at low cost is still in trouble.

The product index standardization of CNC is difficult due to variations in the CNC’s performance obtained from different raw materials, extraction methods, and process parameters. Even the nanocelluloses extracted from the same raw materials can have a certain order impact on the nanocellulose products and affect their properties such as particle morphology, size distribution, mechanical properties, aspect ratio, crystallinity, and other properties. Based on the latest research reports, the wide range mechanical properties of CNCs make it difficult to establish accurate models and performance predictions (120). Therefore, further study is required to develop efficient CNC preparation and characterization methods on a large scale to control its property fluctuations within an acceptable range.

The technical challenges of CNC relate to the lack of designing efficient and precise pretreatments, extraction processes, and surface functionalization. Several toxic chemicals used in these processing steps can disrupt the balance of the ecosystem due to their environmental impacts and non-sustainability issue (121). The hydrophilic nature of CNC greatly limits its usage in hydrophobic polymer matrices. To improve the interfacial compatibility between them, the surface modification of CNC was undertaken by many researchers. However, the modification process influences the physical and chemical properties such as particle morphology, surface microstructure, and crystalline structure, and increases the cost of CNCs further. The effects of CNC modification on the performance of final composites and the complex phases formed with other nanofillers need further study.

CNC’s broader and versatile properties such as sustainability, biodegradability, biocompatibility, and renewability make them bright future materials as they can be obtained from a wide range of natural resources including biomass wastes. Utilization of biomass waste for value-added products could help to have a greener environment by eliminating the polluting wastes. Easy surface modification of CNC helps it to be incorporated
into a variety of materials with different surface chemistry and utilized in a wide range of applications. However, generating the functionalized CNCs by compiling the extraction, chemical functionalization, and polymerization steps into a single process step requires further study.

6. Conclusion

The environmental preservation and sustainability issue increase interest in developing biodegradable and renewable materials. Among the environmental polluting residues, agricultural wastes have been utilized for the production of high-performance CNCs materials. In addition to solving the environmental problems, the development of CNCs from agricultural wastes promotes the local economic development. The biomass sources, executed pretreatments, and extraction processes determine the structural and physicochemical properties of CNCs. Several surface modification techniques have been used to improve the dispersibility of CNCs in hydrophobic polymeric matrices. The excellent desirable properties of CNC such as high mechanical strength, low density, biodegradability, biocompatibility, renewability, high specific surface area, and high thermal stability endow a broad application in biomedicines, nanocomposites, electronics, waste water treatment, food stuffs and packaging, and many more. However, the current production process of CNC requires strong chemicals in multiple steps that result in environmental deterioration, costly process, and restrict commercial-scale exploitation. This review reveals that future researches should be targeted towards developing simplified, cost-effective, and single-step extraction processes with mild reaction conditions without damaging the morphological structure of CNCs.

Acknowledgements

The author would like to thank Dr. Melkie Getnet and Mr. Gemeda Gebino for their valuable comments on this review work.

Disclosure statement

No potential conflict of interest was reported by the author(s).

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