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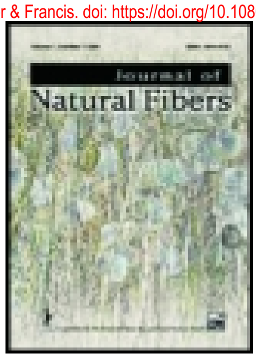

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# Recent advances in isolation, characterization, and potential applications of nanocellulose-based composites: a comprehensive review

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**ABSTRACT**
Cellulose is the most abundant bio-inspired polymer derived from biomass with tremendous promises to expedite the sustainability and green transition. The interesting, fascinating, and applicable properties of nanocellulose-based structures including biocompatibility, low cost, high intrinsic strength, and extraordinary mechanical properties have opened new horizons for their advanced and emerging applications. This comprehensive review aimed to highlight different aspects of cellulose nanomaterials, ranging from preparation, classification, surface modification, nanocomposite fabrication, characterization, and their potential applications in various multifunctional, and high-performance products. This work also reviews the recent approaches applied to modify the surface chemistry of nanocellulose through functionalizing its surface hydroxyl groups to impart advanced desirable properties. Also, emerging applications of CNMs including biosensors, electromagnetic shielding, eco-friendly and sustainable packaging, and bio-medical fields are well demonstrated in this review.

**摘要**
纤维素是从生物质中提取的最丰富的生物启发聚合物, 有巨大的希望加速可持续性和绿色转型. 纳米纤维素基结构的有趣、迷人和适用特性, 包括生物相容性、低成本、高内在强度和非凡的机械性能, 为其先进和新兴应用开辟了新的领域. 这篇综合综述旨在强调纤维素纳米材料的不同方面, 包括制备、分类、表面改性、纳米复合材料制造、表征及其在各种多功能和高性能产品中的潜在应用. 这项工作还回顾了最近应用于通过官能化纳米纤维素表面羟基来修饰其表面化学以赋予高级理想性能的方法. 此外, CNM的新兴应用, 包括生物传感器、电磁屏蔽、环保和可持续包装以及生物医疗领域, 在这篇综述中得到了充分展示.

**KEYWORDS**
Lignocellulosic biomass; Cellulose nanomaterials (CNMs); Cellulose nanocrystals (CNCs); Cellulose nanofibers (CNFs); Nanocomposites; Surface modifications; Biosensors

**关键词**
木质纤维素生物质; 纤维素纳米材料; 纤维素纳米晶体; 纤维素纳米纤维; 纳米复合材料; 表面改性; 生物传感器

## Introduction

Lignocelluloses are the substantial structural component of plant cell walls and are by far the most abundant type of earthly biomass. They have received tremendous attention in association with the current urgent requirements for bio-products, bio-chemicals, and biofuels thanks to their inherent properties such as being highly abundant, low price, availability, and good chemical and biological reactivity (Dutta et al. 2022). They mainly consist of carbohydrates, that is, cellulose (40–60%), hemicelluloses (10–40%), and lignin (15–30%), with minor amounts of extractives, proteins, and inorganic compounds (Fahmy et al. 2017; Djafari Petroudy 2017). Lignocellulose components can

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easily be extracted from both wood cellulose fibers (e.g., spruce, pine, eucalyptus, poplar, etc.) and non-wood biomass, the latter including industrial fibers (e.g., cotton, kenaf, abaca, etc.), underutilized agriculture residues, and agro-food industry from harvesting and pruning operations (e.g., sugarcane bagasse, rice, wheat, and barley straws and empty fruit bunches from oil palm (EFB), etc.) and natural growing plants such as reed, bamboo, esparto grass (Isikgor and Becer 2015; Siró and Plackett 2010).

Reconstruction of low-cost lignocellulosic materials into products with superior functions represents a feasible solution for the improvement of energy security and reduction of greenhouse emissions. Besides different applications of lignocellulosic materials in the conventional pulp and paper industries, there has been a growing interest in the development of promising cellulose-based novel materials in the last decades (Petroudy, Seyed Rahman, and Rudi 2017).

Lignocellulose biomass possesses unique hierarchical structures from millimeter-sized fiber to nano-sized highly ordered crystal units (Habibi, Lucia, and Rojas 2010). Cellulose nanomaterials (CNMs) can widely be assorted into cellulose nanofibers (CNFs) and cellulose nanocrystals (CNCs) or cellulose nanowhiskers (CNWs). CNCs or CNWs are usually elongated rigid needle-shaped or whisker-shaped structures with a diameter of 5–15 nm, a length of several hundred nanometers with a rectangular cross-section. These cellulose nanostructures generally extracted from cellulose and lignocellulosic fibers through mineral acid hydrolyze such as Sulfuric acid ($H_2SO_4$), Hydrochloric acid (HCl), Phosphoric acid ($H_3PO_4$) or combination of them followed by ultrasonication (Eichhorn 2011; Sadeghifar et al. 2011). On the other hand, CNFs have spaghetti-like morphology, a long network structure, and a high aspect ratio with typical dimensions of 20–50 nm in width and up to several microns in length produced by mechanical defibrillation (Isogai, Saito, and Fukuzumi 2011; Saito et al. 2009).

The most widely applied approaches to isolate CNFs are enzymatic treatment and TEMPO-mediated oxidation followed by mechanical liberation (S. Chen et al. 2022). More recently, nano-structured celluloses have widely been applied as a load-bearing agent in different polymer nanocomposites (Abitbol et al. 2016). This intrinsic property is a favorable feature of cellulose nanomaterials thanks to its active hydroxyl groups in order to form the load-bearing percolating structures into the polymer's matrices (Dufresne 2013). Also, inter and intra-particle interactions are able to develop aggregations during the preparation of the nanocomposite due to their high specific surface area and nano-scale size (Eichhorn et al. 2022).

## Preparation of cellulose nanocrystals (CNCs) or cellulose nanowhiskers (CNWs)

The preparation of CNCs from starting raw materials can be grouped into three main steps: (1) purification (2) acid hydrolysis and (3) mild mechanical treatment (L. Zhang et al. 2022). The former is generally employed to remove other constituents such as non-cellulosic substances than cellulose fibers. The acid hydrolysis step involves the hydrolysis of purified cellulose into nanocrystals or nanowhiskers components as shown in the following illustration (Figure 1). Different lignocellulose-containing sources (e.g., agricultural by-products, wood, and non-wood) are initially subjected to different pretreatment processes which reduces time by allowing chemical accessibility in the second step (Wei et al. 2018). Disordered or para-crystalline regions of cellulose fibers are preferentially hydrolyzed through the reaction with strong mineral acids, whereas crystalline regions that have a higher resistance to acid attack remain intact (Habibi, Lucia, and Rojas 2010). The obtained product consists of the well-ordered crystalline region which varies with regard to its starting raw materials, type of acid, hydrolysis time, and reaction temperature. Sulfuric acid and hydrochloric acid are the commonly applied acids to afford the extraction of CNCs, but other acids such as hydrobromic (Sadeghifar et al. 2011; Sucaldito and Camacho 2017), phosphoric (Camarero Espinosa et al. 2013; Tang et al. 2014), and phosphotungstic acid (Lu et al. 2016), or even combination of these acids (Amin et al. 2022) were also proposed for such purpose. The morphology and crystallinity of CNCs are different from their pristine cellulose fibers. In general, acid hydrolysis of native cellulose enforces a rapid decrease in its degree of polymerization (DP), to the so-called level-off DP (LODP). The DP

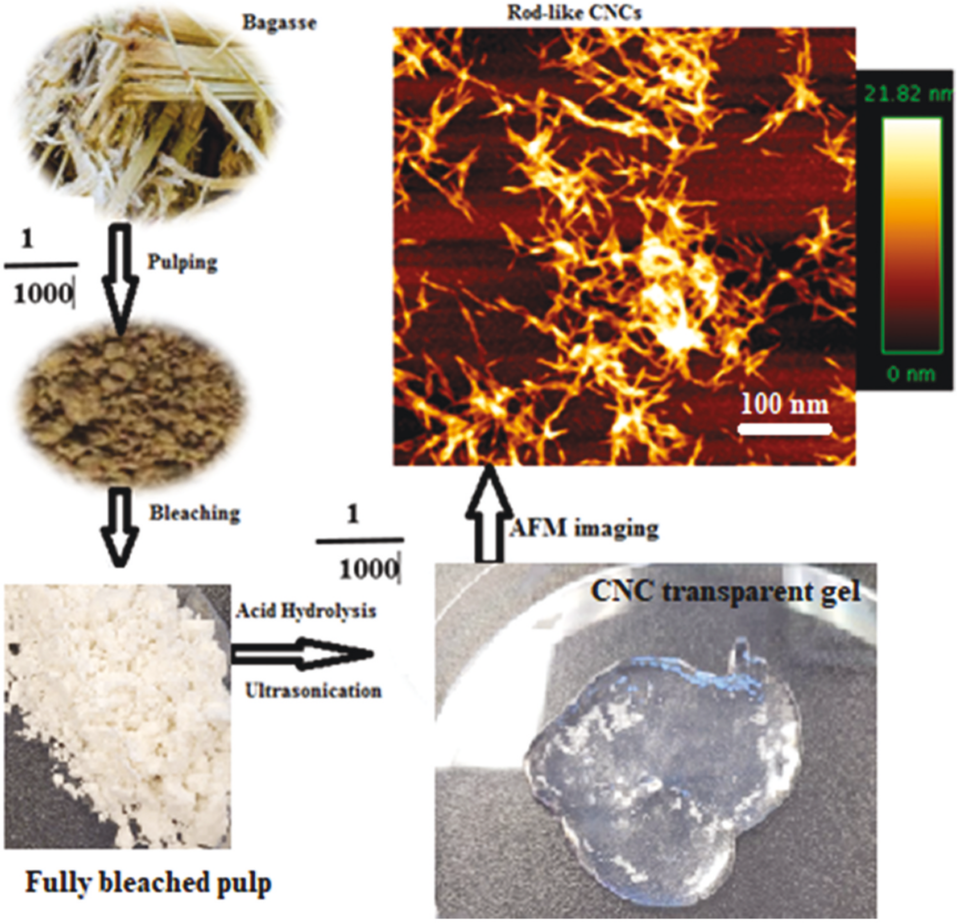

**Figure 1.** The hierarchy illustration for preparation of CNCs from bagasse.

subsequently decreases much more slowly, even during prolonged hydrolysis times (Habibi, Lucia, and Rojas 2010). In the third step employing high-intensity ultrasonication is essentially required for the prevention of the extracted nanocrystals to agglomerate (Azadbakht et al. 2016). The hierarchical approach for the preparation of CNCs from bagasse is shown in Figure 1.

Recently, the preparation of lignin-rich CNCs (LCNCs) follows the same approach as conventional CNCs especially employing well-documented acid hydrolysis (Agarwal et al. 2018; Wei et al. 2018). The distinctive factor is the extent of delignification of the various sourced cellulose fibers. Whereas CNCs employ lignin-free pulps, the LCNCs use unbleached or partially bleached pulps containing residual lignin (Ojala, Sirviö, and Liimatainen 2018). It is worth noting that the higher the lignin content, the more savings from the pulp bleaching steps resulting in easier processing, and low-cost material with more possibility of easy industrial-scale utilization, and commercialization (Petroudy, Seyed Rahman, and Vatankhah 2021). However, unbleached cellulose fibers offer a significant improvement in CNCs quality and process effectiveness including total energy-saving and diminishing pollutants loads from bleaching treatments. Moreover, the process for the preparation of LCNCs using a combination of maleic acid hydrolysis and mechanical fibrillation has been reported (Bian et al. 2017). The pulps were hydrolyzed using 60 wt% maleic acid solution at 120°C for 120 min under ambient pressure. The resultant mixture was later centrifuged, dialyzed, and centrifuged again to obtain the LCNCs. The lignin-rich CNCs can be reported as a promising functional bionanomaterial for numerous applications such as packaging, effective adsorbents for dyes, and reinforcing agents to improve the physical, and thermal properties of polymer nanocomposites (Geng et al. 2018).

## Applications of cellulose nanocrystals

In the most recent decade, the application of renewable materials on the industrial scale is becoming impellent owing to the growing request for alternatives to scarce and unrenewable traditional supplies. In this regard, CNCs extracted from native and recovered cellulose fibers via strong acid hydrolysis have received great attention from the materials community.

## CNs as sustainable strength-enhancer for polymers

The application of CNCs as an extraordinary, and bio-inspired nano-reinforcer in polymer nanocomposites has been well demonstrated (Chen et al. 2019; Miao and Hamad 2019). CNCs with high aspect ratio and surface area have been incorporated as strengthening agents into a wide range of polymer matrices such as poly(lactic acid) (Karkhanis et al. 2018), polyamide (Nicharat et al. 2015), poly(oxyethylene)(Azizi Samir et al. 2004), polypropylene(Gray et al. 2018), poly(vinyl alcohol) (Van Nguyen and Lee 2022), starch (Liu et al. 2014), natural rubber (Cao et al. 2018), and poly(methyl methacrylate) (Ni et al. 2018). It is reported that the incorporation of CNCs into polymers would result in the formation of a percolation network, connecting well-dispersed CNCs by hydrogen bonds, and reinforcing polymer (Favier et al. 1995). In general, the performance of CNCs as nano-reinforcing agents is governed by different factors such as CNCs' characteristics, morphology, and dispersion in the polymer matrices. In this regard, extensive works have been conducted to modify the surface of CNCs to improve the dispersion in polymer matrices (Ansari et al. 2015; Shan et al. 2019; Tan et al. 2015). Kupka et al. modified CNCs by grafting poly(ethylene glycol) on the CNC surface (Kupka et al. 2018) then they well-incorporated modified CNCs into polyurethane. Their results confirmed strong enhancements in mechanical properties of prepared nanocomposites up to 50% due to an improvement in cellulose nanocrystal dispersion in polyurethane. They have also confirmed an improvement in the glass transition temperature ($T_g$(°C)) of nanocomposites. The addition of just 1.0 wt.% of cellulose nanocrystal increased the $Tg$ by 5°C, with respect to the pure polyurethane. In another work, Geng et al. reported well-dispersed cellulose nanocrystals in hydrophobic poly(vinyl acetate) and poly(vinyl acetate)-Poly(lactic acid) using in situ polymerization Figure 2(b,c) compared to PLA/PVAc blends with no CNCs (Figure 2a). The interaction between the CNCs and poly(vinyl acetate) was improved by the formation of crosslinking after polymerization, and this resulted in higher

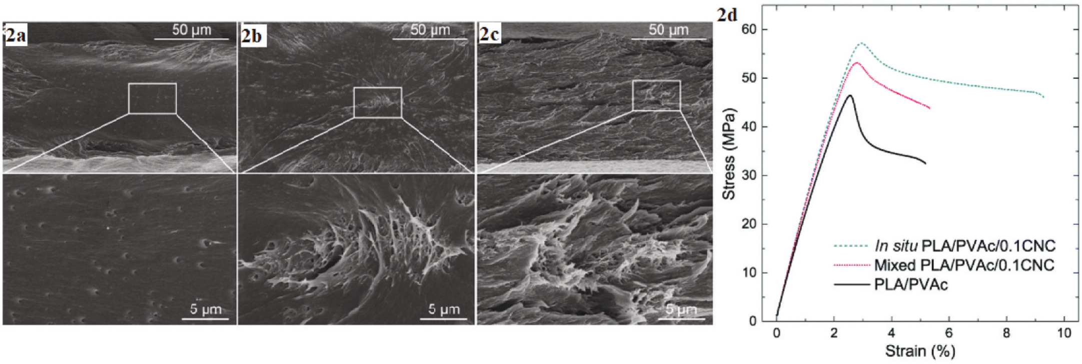

**Figure 2.** SEM micrographs of the fracture surfaces of the (a) PLA/PVAc (weight ratio 90/10), (b) mixed PLA/PVAc/0.1CNC and (c) *in-situ* PLA/PVAc/0.1CNC after tensile testing. (d) Stress – strain curves of PLA/PVAc/CNC nanocomposites (reproduced with permission from (Geng et al. 2018).

mechanical properties of nanocomposites. They have also demonstrated that the *in-situ* approach results in a remarkedly more homogeneous and ductile fracture surface after tensile testing (Figure 2d) (Geng et al. 2018).

## Pulp and paper industries

The pulp and paper industries are one of the largest CNCs consumers in the world dominated by North America, European, and eastern Asian countries (Samyn et al. 2018). Papers are considered as thin layer of pulp fibers in contact with each other owing to the formation of hydrogen bonding between hydroxyl groups located on the surface of CNCs (Petroudy et al. 2019). Generally, typical raw materials for making papers include softwoods (low-density long fibers), hardwoods (high-density short fibers), non-wood-based materials such as bagasse, bamboo, kenaf, reed, straw, and grasses and recovered or recycled paper (Jafari-Petroudy, Resalati, and Rezayati-Charani 2011). High strength and stiffness are considered the main promising features of CNCs which can improve the overall properties of paper-based materials in either wet or dry conditions (Osong, Norgren, and Engstrand 2016). In this regard, CNCs have been applied as a bio-inspired and distinctive additive for pulp in the papermaking process to import high optical transparency to paper.

## Sustainable and eco-friendly food packaging

Global environmental interest in superseding petroleum-based traditional polymers with biodegradable packaging materials has motivated researchers and industries in introducing new natural polymers. Despite great advances in biopolymers, the application of bio-based polymers for food packaging still suffers from low mechanical characteristics and poor barrier performance against oxygen, water, water vapor, and light (Petroudy et al. 2019). In this regard, the incorporation of nanosized fillers such as CNCs is a well-considered potential solution (Andrade et al. 2022; El Achaby et al. 2017). Superior mechanical characteristics, highly crystalline structure, low cost, biocompatible and biodegradable features, make CNCs a promising bio-based material in the food packaging industry (Wang et al. 2018). The addition of CNCs improved the overall barrier properties required for food packaging by reducing water vapor and oxygen permeability in host polymer films. More recently, multifunctional cellulose nanocrystal films were prepared through a facile method by applying to cross-link between metal ions ($Al^{3+}$ and $Fe^{3+}$) and –O–SO3– groups on the CNC surface (Chen et al. 2022). These films showed high UV absorption properties, the excellent barriers to oxygen, water vapor, oil, and grease. It is demonstrated that these films with fire self-extinguishment properties are promising for the next generations of the sustainable packaging. In particular, the use of CNC as biobased nanofillers has been widely studied to improve the PLA barrier properties. For instance, Fortunati et al conducted a study on the application of modified and pristine CNCs on barrier properties and migration behavior of PLA-based nanocomposites (Fortunati et al. 2013, 2014). They reported that surfactant-modified CNCs dispersed uniformly in PLA and significantly reduced water permeability. They also observed little tortuosity in PLA nanocomposites due to the incorporation of CNCs. The combination of surfactant-modified CNCs with silver particles in improving the barrier properties of PLA was also reported (Fortunati et al. 2014). In addition to higher barrier properties, the introduction of silver particles and CNCs resulted in antimicrobial activity in the resultant nanobiocomposite films (Figure 3). These optical images well explore the quite homogeneous distribution of Ag nanoparticles in the nanocomposite films, which did not appear to be affected by the presence of CNCs. However, due to the lack of enough information on the health effects of silver nanoparticles, their application cannot commercialize at this time (Espino-Pérez et al. 2018). Hybrid CNCs decorated with silver nanoparticles have previously been prepared (Errokh et al. 2018). This research group reported that the hybrid CNC-Ag nanoparticles showed good bactericidal properties against both Gram+ and Gram− bacteria. Interestingly, the amount of Ag released from the hybrid films was 1.6 ppb 48 h which is much lower than the recommended permissible limit by US National Institute for

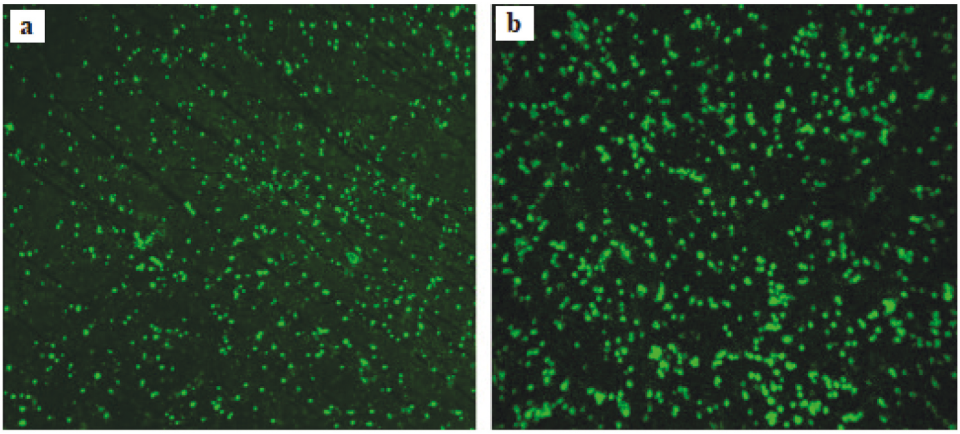

**Figure 3.** Confocal images of (a) PLA-1ag (scan area 100 × 100 μm$^2$), and (b) PLA/1-sCNC-1ag (scan area 130 × 130 μm$^2$). (reproduced with permission from (Fortunati et al. 2014)).

Occupational Safety and Health (NIOSH). Such nanocomposite films would be able to contribute promising potential applications in coating and biomedical sectors with enhanced antibacterial activity. Dhar et al. studied the effects of CNCs in Poly (3-hydroxybutyrate) for food packaging. They found that the oxygen transmission rate decreased significantly due to crystal nucleation and tortuosity (Dhar et al. 2016). In a similar approach, bio-nanocomposites films of cassava starch plasticized with sucrose, inverted sugar, and reinforced by CNCs were prepared by solution casting. The high crystallinity of CNCs not only improved the water-resistance properties of thin films, but also strengthen the thin films (da Silva, Pereira, and Druzian 2012).

In addition to incorporating CNCs into different polymers, the effect of a thin layer of crystalline CNCs, applied as a coating or by lamination in enhancing the barrier properties of polymers has been studied. In a study conducted by Herrera, two types of thin films made of alternate layers of poly (allylamine hydrochloride) and CNCs, or a single layer of poly(allylamine hydrochloride) coated with multilayers of CNCs prepared using the spin-coating method. Their results claimed the low permeability for $N_2$ and $O_2$ and gas selectivity for some gas combinations in thin films. This shows that the gas molecules diffused easier through the film with higher polymer content and lower tortuosity (Herrera, Mathew, and Oksman 2014).

## Bio-medical and hygienic products

Cellulose nanocrystals with biodegradable, nontoxic, low cytotoxicity to a range of animal and human cell types, and biocompatible nature is a research topic in pharmaceutical application over the last two decades. In this regard, CNCs serve as additives in different fields such as wound dressing, drug delivery, tissue engineering, and medical implants (Ju et al. 2020; Khadivi et al. 2019; Tang et al. 2019). The application of CNCs in combination with silver nanoparticles in expediting the healing process of diabetic wounds has been evaluated in a work by Singla's research group. Bamboo cellulose nanocrystals were used as matrices and silver nanoparticles were impregnated via *in-situ* treatment. Their results exhibited that CNCs improved the re-epithelialization and collagen deposition in wounded skin (Singla et al. 2017). In another approach, CNCs were used as a reinforcement agent in a gelatin-based hydrogel. Excellent pH sensitivity was reported for hydrogels due to the network formed between CNCs and gelatin. The provided hydrogel with a controlled drug delivery function was employed as a carrier for theophylline (Ooi, Ahmad, and Amin 2016). The formation of the cross-linked network between CNCs, gelatin, and sodium alginate provides a porous scaffold hastening skin regeneration in rats (Figure 4a). Thanks to hydrogen bonding, the incorporation of 0.5% CNCs

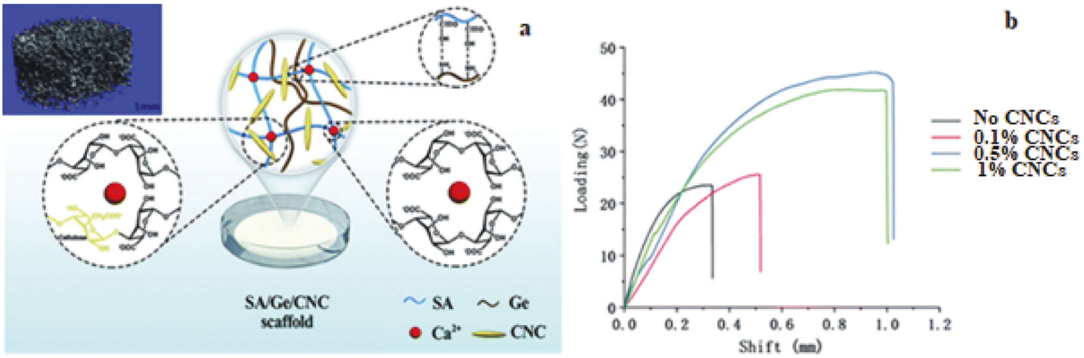

**Figure 4.** (a) Schematic diagram of hydrogen bonds formed between CNCs, gelatin, and sodium alginate. (b) Stress–strain curves of hydrogels as a function of CNC loadings. (reproduced with permission from (Shan et al. 2019)).

yielded an increase in interactions between hydrogel components, let to formation entangled network, and resulted in a significant improvement in tensile strength. Although, it should be mentioned that there was not much significant difference between the tensile strength at 0.5% and 1% of CNC loading. (Shan et al. 2019)(Figure 4b).

Another interesting field, in which CNCs have received huge interest is tissue engineering. The formation of the porous scaffold with multiscale open-pore architecture promoting cell attachment and growth is a capability of CNCs in preparing hydrogel scaffolds. In this regard, a composite scaffold made of chitosan-g-D, L-lactic acid, and CNCs have been tested for application in tissue engineering. An increase in porosity, degradability, drug release property, and cell viability were reported for composite scaffolds reinforced by CNCs (Ko et al. 2018).

## Synthesis of cellulose nanofibers and lignocellulosic nanofibers (CNFs and LCNFs)

The CNFs and LCNFs are usually prepared from the starting bleached and unbleached cellulose fibers through a mechanical process. In this process, a higher shear force is generated which results in the extraction of CNFs and LCNFs along the longitudinal direction from the cellulose fibers as shown in Figure 5. The most commonly used approaches to isolate CNFs and LCNFs from their starting materials are enzymatic pretreatments (cellulase, xylanase), chemical pretreatments (TEMPO oxidation, carboxymethylation) followed by mechanical defibrillation. The final product after the delamination process is gel-like CNFs and LCNFs dispersion in water. CNFs have spaghetti-like morphology with typical dimensions of 20–50 nm in width and up to several microns in length. Four frequently and well documented used mechanical approaches for the production of both CNFs and LCNFs are ultrafine supergrinders or microgrinders, high-pressure homogenizers (HPHs), and high-pressure microfluidizers (HPFs), and Twin-screw extruders (TSEs). However, the mechanical process is usually associated with high-energy consumption, which causes cellulose fiber delamination. The chemical pretreatment process helps in reducing energy consumption up to 98%. Optimum selection of these mechanical techniques depends on some decisive factors including type, morphological, and chemical characteristics of starting raw materials, the fibrillation severity, final product specifications, and end-use of prepared CNFs and LCNFs. Commercial production of CNFs and LCNFs is now encountering serious challenges such as high energy demand and high investment costs (Petroudy, Seyed Rahman, and Vatankhah 2021). To conquer these obstacles, various solutions include the use of cheaper raw materials (e.g., underutilized agricultural residues) (Pennells et al. 2020), pulp and paper sludges (Adu et al. 2018; Jonoobi, Mathew, and Oksman 2012), unbleached cellulose fibers (Petroudy et al. 2019), enzymatic (Henriksson et al. 2007; Petroudy et al. 2015), and chemical pretreatments (Petroudy, Seyed Rahman, and Rasooly Garmaroody 2018; Saito et al. 2006) have extensively been proposed.

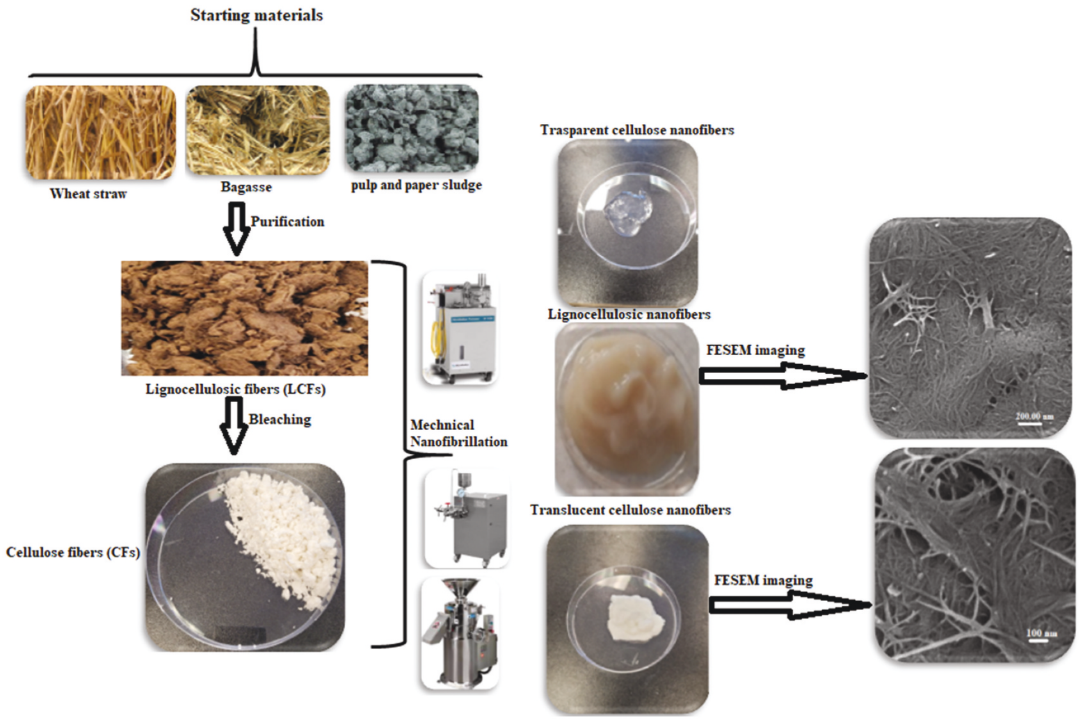

**Figure 5.** The up-to-down approach for preparation of CNFs and LCNFs from different underutilized lignocellulosic fibers.

Interestingly, it is shown that Water hyacinth would be an excellent and sustainable candidate to be applied for the large-scale production of CNFs to maximize its capability for advanced applications compared to wood or other lignocellulosic fiber owning to a lower requirement for chemical treatments (Supachok Tanpichai, Biswas et al. 2019; Tanpichai et al. 2021). It is worthy mentioning that, its nanocellulose-based hydrogels can be applied to Superabsorbent fibers with swelling ratio of 900% (Tanpichai, Phoothong, and Boonmahitthisud 2022). It is concluded that by applying chemical pretreatment through TEMPO-mediated oxidation, the specific energy consumption can interestingly be lowered below 1.9 Wh/Kg while procuring a homogeneous transparent hydrogel with a fibril diameter of 3–4 nm and a length of a few micrometers (Isogai 2018). Carboxymethylation pretreatment is also named one of the most prevalent pretreatment routes for the production of CNFs, where anionic carboxylate groups are introduced onto cellulose. This pretreatment also results in CNFs with low energy consumption. Periodate – chlorite as a sequential regio-selective oxidation pretreatment has recently been applied for the production of CNFs. This pretreatment reduces the hydrogen bond by increasing the anionic charge density of cellulose fiber surfaces, thereby decreasing the required mechanical energy for the liberation of cellulosic fibers (Petroudy, Seyed Rahman, and Vatankhah 2021). Figure 5 presents the top-down approach for the preparation of CNFs and LCNFs from different underutilized lignocellulosic fibers.

## Applications of CNFs in different nanocomposites

Recent trends in different industries shift many applications to the nanoscale level with special emphasis on the advantages of nanotechnology for the improvement of functional properties of nanocomposites. On the other hand, serious concerns regarding both economic and environmental sustainability have led to the development of more sustainable alternatives for petroleum-based materials. In this case, biodegradable and bio-based nanocomposites, with the application of synthetic and bio-based materials are an effective solution for replacing conventional synthetic and non-biodegradable materials in diverse sectors.

CNFs and LCFs with low cost, tunable surface properties, and enhanced mechanical characteristics have been considered as potential nanoscale reinforcer agents or matrices for various applications such as sustainable packaging, electromagnetic shielding, energy storage devices, bio-imaging, optoelectronic conversion, drug delivery and biomedical materials, protective coatings.

## Sustainable reinforcing agent

The most important characteristics of CNFs and LCNFs are their high strength, high surface area, and low density compared to other natural and synthetic nanofibers besides their biodegradability and biocompatibility. These features make CNFs and LCNFs as promising as sustainable reinforcement nanofibers for the development of high-performance nanocomposites. Regarding the presence of a high number of hydroxyl groups on the surface of CNFs and LCNFs, water-soluble polymers are the most promising medium for preservation of CNFs and LCNFs dispersion and nanocomposites manufacturing (Sharma et al. 2019). Despite the overall compatibility between CNFs and LCNFs and water-soluble polymers, CNFs and LCNFs tend to form irregular aggregates due to their high crystallinity and strong intermolecular hydrogen bonds. Hence, in order to exploit the maximum potential of CNFs and LCNFs as reinforcement, uniform dispersion of hydrophilic CNFs and LCNFs in different polymer matrices was extensively investigated via different surface modification treatments such as acetylation, TEMPO modification, esterification, polymer grafting (Iwamoto et al. 2019; Liu et al. 2019; Ávila Ramírez et al. 2019). The incorporation of modified and unmodified CNFs and LCNFs in improving mechanical, thermal, rheological, and barrier properties of different polymers such as poly(lactic acid), polypropylene, polyethylene, polyethylene glycol, and polyvinyl alcohol have been extensively investigated (Cailloux et al. 2019; Jamaluddin et al. 2019). It was reported that in addition to non-uniform dispersion CNFs and LCNFs with an aspect ratio smaller than 10 cannot improve the mechanical characteristics of nanocomposites. Only CNFs and LCNFs with aspect ratios greater than 50 can ensure a noticeable reinforcement effect. In this regard, Wang et al. reported superior mechanical characteristics in polypropylene (PP) nanocomposites reinforced by CNFs with different degrees of substitution (Wang et al. 2019). Alkenyl succinic anhydride (ASA) was employed to improve the compatibility between CNFs and polymer and resulting in a noticeable improvement in the rheological properties of nanocomposites. The improvement in rheological characteristics was more likely due to high integration between ASA-modified CNFs and PP (Long Wang et al. 2018). Another environmentally friendly approach was implemented by Kaushik et al., for the production of thermoplastic starch reinforced by alkaline steam-exploded CNFs (Kaushik, Singh, and Verma 2010). In addition to more uniform dispersion, smaller fiber diameter was reported through chemical treatment, which in turn, resulted in an improvement of 195% in mechanical properties of nanocomposites over pure thermoplastic starch. Load bearing is another application that CNFs and LCNFs play in nanocomposite hydrogels with potential applications in different fields. In this regard, CNFs and LCNFs can be dispersed in a monomer solution and then incorporated into a host polymer matrix network, expecting to increase the stretch-ability, keep the stiffness of the hydrogel networks, and achieve better mechanical performance. Niu et al. proposed a facile method of synthesizing physically cross-linked polyacrylamide nanocomposite hydrogels reinforced with CNFs and ferric ions using radical polymerization (Niu et al. 2018). The remarkable mechanical properties with high stiffness and toughness, quick recovery, and healing facilities were observed in hydrogel due to the distinctive roles of dual physical crosslinks. In another study, TEMPO-oxidized CNFs were incorporated into chitosan to prepare injectable hydrogel with biocompatibility and degradability (Nguyen et al. 2018). As sol-gel transition at body temperature is an important specification for hydrogel with potential application in the biomedical sector, the sol-gel transition behavior of hydrogel was studied and the addition of CNFs resulted in faster gelation time and increased porosity. LCNFs has recently been used to strengthen polypropylene nanocomposites (Ninomiya et al. 2018). It is demonstrated that LCNFs have played a crucial role to prepare high-strength nanocomposites. Lignocellulosic nanofiber was more recently used to prepare reinforced PLA-based composites for 3D printing (Q. Zhang et al. 2022). In this study, LCNFs with different lignin contents (0.2–8.5%) were mixed with PLA and were 3D-printed to fabricate printable nanofilaments. The LCNFs enhanced the thermal stability and mechanical strength of the resultant printable nanofilament. These unique characteristics of LCNFs are promising as ideal nano-reinforcer for common fused deposition modeling (FDM) 3D printers.

## Electromagnetic shielding

The electromagnetic waves produced by electric devices strongly affect the proper performance of other electrical devices. Furthermore, radiation pollution harms the network of veins and nerves in high-risk organs such as the eyes more likely owning to thermal interactions. In order to eliminate or minimize the potential hazards to human beings and to protect the sensitive circuits from the interference of electromagnetic waves, the importance of application of electromagnetic shields in a wide range of frequencies has recently drawn remarkedly attention (Mondal et al. 2017). New emerging functional synthetic nanomaterials, such as graphene, CNTs, and metal nanoparticles, are confined owning to weak interfacial interactions, resulting in unsatisfied mechanical properties. Thus, the current approach of combining these nanomaterials with CNFs in an endeavor to conquer structural and mechanical deficiencies have been remarkedly attracted. In view of EMI shielding performance, the significant interactions between CNFs and functional conductive nanofillers can results in a strong integration with each other. The nature of electromagnetic shields requires their flexibility, lightweight, and sustainability to serve in different sectors (Zhang et al. 2019). Therefore, the organic polymers with relatively high conductivity will be overtaking metals (Tantawy et al. 2013), if their rigidity and brittleness are completely addressed. In this case, CNFs are currently gaining significant attention as extraordinary nanofibers for polymer composites thanks to their outstanding mechanical characteristics (Yang et al. 2017a). Electromagnetic shielding materials containing CNFs have considerably received interest since the 1970s due to their superior mechanical characteristics, controllable synthesis process, and low density (Omura et al. 2019). In spite of the extensive number of studies on the application of CNFs as electromagnetic shields, its potential application is still limited due to the difficulty in achieving uniform dispersion, and weak interfacial strength between composite components (Zhang et al. 2019). Therefore, different methods have been investigated to achieve a uniform distribution of CNFs in the host polymer matrices (Zeng et al. 2022). Oxidative polymerization is the most popular method in synthesizing CNFs conductive nanocomposites to improve its electromagnetic shielding efficiency. An oxidant agent is incorporated into well-dispersed CNFs slurry in order to initiate monomer polymerization. The optimum interaction between polymer and CNFs surfaces was improved via multiple hydrogen bonding between the −NH and−OH groups of Polyaniline (PANI) and the CNFs surface (Omura et al. 2019). They have successfully prepared an organic thin nanopaper of CNFs/PANI using the oxidative polymerization approach followed by vacuum filtration and washing (Figure 6). In this study, the freeze-dried CNF films were coated by PANI doped with (±)-10-camphorsulfonic acid nanohybrid, and ammonium persulfate (APS) was used as the oxidizing agent.

Cao's group fabricated an ultrathin and highly flexible $Ti_3C_2T_x$ (d-$Ti_3C_2T_x$, MXene)/CNFs composite paper with a nacre-like lamellar structure via a vacuum-filtration-induced self-assembly process (W.-T. Cao et al. 2018). They introduced an ultrathin paper with a minimum thickness of 47 μm with the potential application in wearable devices, and robot joints as they observed high tensile strength (up to 135.4 MPa), fracture strain (up to 16.7%), folding endurance, and specific electromagnetic interference shielding. Another research group in 2017 adopted a vacuum-assisted filtration followed by hydroiodic acid (HI) reduction method to achieve an ultrathin highly aligned reduced graphene oxide/CNFs nanocomposite film (Tang et al. 2014). They reported an outstanding electromagnetic interference shielding and notable electrical conductivity attributed to the uniform dispersion and self-alignment into the layered structure of reduced graphene oxide.

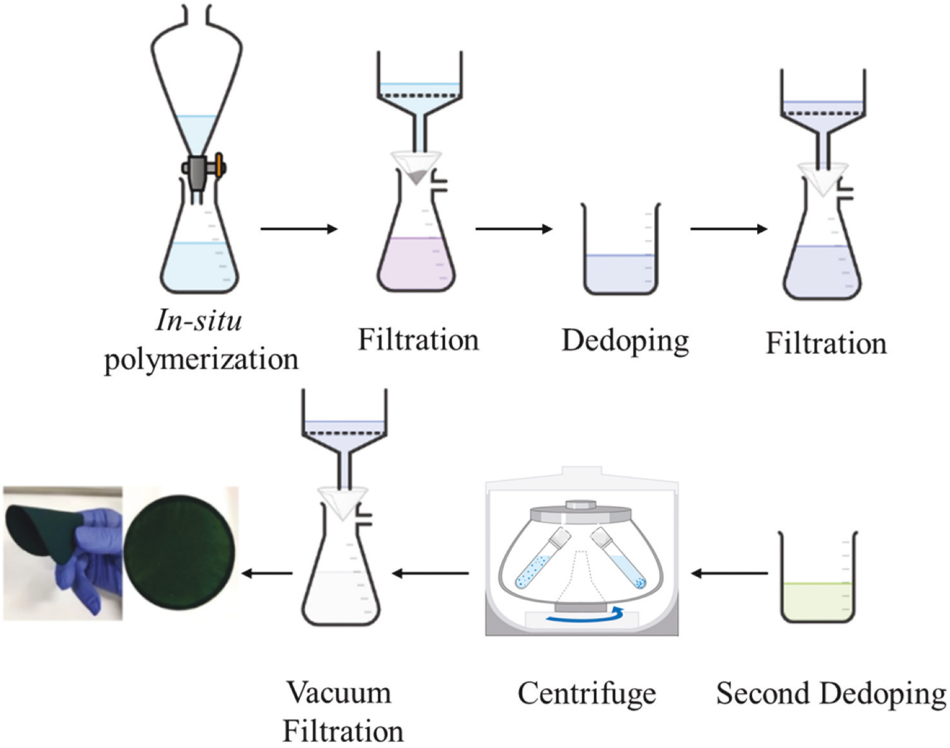

**Figure 6.** Synthesis of polymer coated CNF paper. Reproduced with the permission from (Omura et al. 2019).

## Sustainable and eco-friendly packaging

Food safety and its quality are substantially dependent on packaging materials. These crucial factors play an important role, mainly due to increased demands in terms of product safety, shelf-life extension, cost efficiency, environmental issues, and consumer convenience. A strong public concern for environmental protection triggers the use of sustainable food packaging as an alternative to materials with a nonrenewable basis. However, the application of biopolymers as food packaging materials is still suffering from some drawbacks such as low mechanical, thermal, and barrier properties compared to conventional non-biodegradable materials. Therefore, a growing number of investigations have been directed toward the development of nanocomposite films with acceptable mechanical, thermal, and barrier characteristics for serving as food packaging products (Ghaderi et al. 2014; Youssef and El-Sayed 2018). Among different bio-based materials with a potential application in food packaging, CNFs have received substantial interest as a nanoscale reinforcing agent to improve mechanical characteristics, barrier properties, and glass transition temperature of nanocomposite films (Tanpichai et al. 2019). In addition, the use of CNFs can not only improve the food shelf life, but also it can enhance food quality as CNFs can hold some active substances, such as antioxidants and antimicrobials (Khan et al. 2014). The barrier properties, as well as mechanical characteristics, are considerably affected by the nanofiber's size distribution or agglomeration in the host polymer matrices, and the formation of CNF aggregates lowers the CNF surface area and weakens interfacial bonding between nanofiber and polymer matrix since they do not reflect the properties of the individual nanofiber (Patra et al. 2018). Different approaches have been addressed to improve the dispersion of CNFs as a strength enhancer into the host polymer matrices (Deng, Jung, and Zhao 2017; Samadani, Behzad, and Enayati 2019). In the food packaging sector, CNFs were either applied as the main component of nanocomposite films or as a strength enhancer. Generally, CNF films are formed

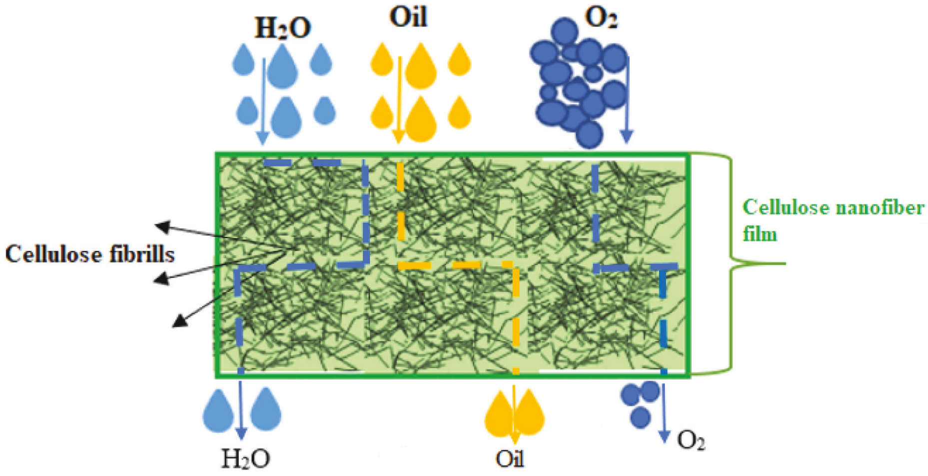

Figure 7. Barrier performance of nanocomposite films containing CNFs studied for packaging products.

from CNFs aqueous suspensions by either solvent casting or drying under vacuum, or filtration followed by drying. The low porosity of CNF films facilitates oxygen diffusion, and adequate thickness results in the impermeability of thin films. However, the oxygen transmission rate (OTR) of films greatly depends on the CNFs production methods, and film thickness (Aulin, Gällstedt, and Lindström 2010) (Figure 7). In the case of water vapor permeability, nanocomposite films containing CNFs lose much of their ability since CNFs are inherently sensitive to the presence of both gaseous and liquid water (Hubbe et al. 2017). It was reported that virgin CNF films typically exhibit an OTR of less than 1 $cc/m^2/day$ under ideal conditions (Nguyen et al. 2018) and the application of CNFs chemical modification or incorporation CNFs into different hydrophobic polymers has been speculated as key solutions to tune the barrier properties of CNF films even at high humidity but has not been fully effective.

Niu et al. synthesized multifunctional composite films from CNFs and (PVA) (Niu et al. 2018). Prior to the incorporation of CNFs into the polymer matrix, TEMPO-oxidized CNFs were prepared in the heterogeneous phase with benzophenone, diisocyanate, and epoxidized soybean oil via esterification reactions. In this regard, high UV blocking performance, especially in the UVB region was observed. As demand for the safety and quality of food packaging products increases, there has been a lot of interest in using active packaging to extend the shelf-life of food products and preserve food quality, safety, and integrity. Active packaging is normally designed to provide a deliberate interaction between packaging materials with food and/or its direct environment. In one effort, Chargot's research group synthesized a CNF nanocomposite films by incorporating low viscosity chitosan into carrot CNFs to improve the nanocomposite hydrophobicity and antimicrobial properties (Szymańska-Chargot et al. 2019). Different concentration of chitosan was added to CNFs aqueous suspension and nanocomposite films were formed via vacuum filtration followed by air-drying method (Figure 8). The higher barrier properties of nanocomposite films compared to pure CNF films were more likely attributed to physical interactions between CNFs and chitosan rather than hydrogen bonding. They observed a uniform dispersion of chitosan in the CNF matrix, however, at higher chitosan content thin-film transparency decreased as a result of CNF aggregates.

## Superabsorbent fibers

Superabsorbent polymers (SAPs) have been extensively applied in various applications including hygienic products such as diapers, napkins, incontinence pads, Feminine hygiene products, drug delivery systems, catalysis, biosensing, wastewater treatments, water blocking tapes, and horticulture

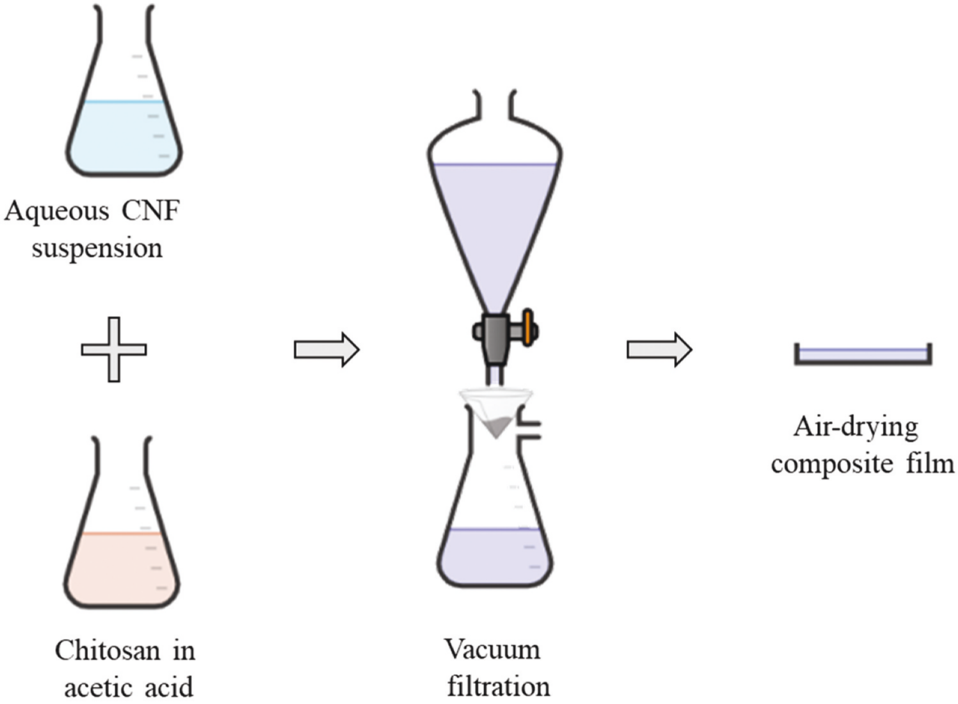

Figure 8. Synthesis of CNF-chitosan paper. Reproduced with the permission from Springer (Szymańska-Chargot et al. 2019).

(Petroudy, Seyed Rahman, and Rasooly Garmaroody 2018). Most of the commercial SAPs are petroleum-based and are synthetically produced from Acrylic acid (A.A), its salt, acrylamide, and cross-linked Sodium polyacrylate (SPA) gels. Depending on the ionic strength of the fluid, these SAPs are able to absorb many times their own weight in aqueous liquids, in the case of tap water, about 200 times (Petroudy et al. 2021). The tendency to find a sustainable product replacing synthetics is more overwhelming owning to the poor degradability and biocompatibility of the former. Hitherward, different kinds of polymers have been applied to fabricate three-dimensional SAPs, among them the bio-inspired available resources such as polysaccharide-based have explored particular advantages and drawn considerable attention. More recently, CNFs thanks to their capability to form percolated three-dimensional architecture connected by hydrogen bonds have gained increasing attention (Petroudy et al. 2021; Zhang et al. 2022). Different water absorption rates have been reported for CNFs-based superabsorbent fibers. The best swelling capacity of 200 gr/gr was addressed along with addition of 10% TEMPO-mediated CNFs (Petroudy, Seyed Rahman, and Rasooly Garmaroody 2018). In this regard, Wang et al., synthetized a hierarchically porous structure from CNFs with ultra-low density (2.7 mg·cm$^{-3}$). The superhydrophobic CNFs with contact angle of 150.5° exhibited outstanding oil absorption (126.8–320.4 g·g$^{-1}$) and oil-water separation performance (Zhang et al. 2022). It is worth noting that, chemically (Hexa methyl diamine (HMDA)) crosslinked TEMPO-oxidized nanocellulose SAPs biodegraded to a higher extent (~56%) compared to neat nanocellulose SAP (~41%) in soil under cellulase-assisted conditions.

## Biosensors

Recently, comprehensive research works has been performed to the development of enhanced sensitivity, specificity, detection time, and low-cost biosensors as substitutes for traditional sensing systems (Baptista 2013). The attractiveness of CNFs as functional materials for bio-sensing areas relies not only on their ability to act as an efficient and stabilizing matrix for bio-sensing elements, but also on their large surface area, controlled morphology and structure, and biocompatibility (Baptista 2013). The special chemical structure of CNFs enables them to be applied for the immobilization of bioactive molecules. Enzymes can be linked with CNFs through covalent linkages, salt bridges, and physical inclusion complexes (T. G. Kim and Park 2006). In addition, different non-covalent surface biosen treatments such as surfactants adsorption, sulfonation, silylation, amidation, TEMPO-mediated oxidation, esterification, etherification, urethanization, and polymer grafting can preferably alter CNFs structure to specific functional groups. An innovative and efficient approach was proposed by Weishaupt et al. (Weishaupt et al. 2015). They reported on the use of a two-step method for the stable immobilization of small peptides and enzymes onto TEMPO-oxidized CNFs. The introduction of negative charge on the CNFs surface through TEMPO oxidation implemented the absorption of biomolecules and cross-linking reaction with 1-ethyl-3-[3-(dimethylamino)propyl] carbodiimide (EDAC) and glutaraldehyde (Ga) chemistry. A different approach was reported by Ma and Ramakrishna. In which, they oxidized the CNFs membrane by $NaIO_4$ for activation of hydroxyl groups in the membrane and immobilized the protein A/G covalently on the membrane (Figure 9). The membrane was then capable of specifically capturing IgG molecules. No morphological change was observed after the modification (Ma and Ramakrishna 2008).

In particular, thanks to the high number of carboxylic groups on the CNFs surface, they are capable of determination of the binding and rate constant for small molecules. In a study conducted by Sathishkumar 's research group, the immobilization of laccase on the cellulose nanofiber was conducted using CNF dispersed in sodium acetate buffer and mixed with laccase. The results showed that immobilized laccase had an efficient catalytic ability (Figure 10). The high simulated dye effluent (SDE) de-coloration results revealed the potential application of laccase immobilized cellulose nanofiber in the bioremediation of textile dyes (Sathishkumar et al. 2014).

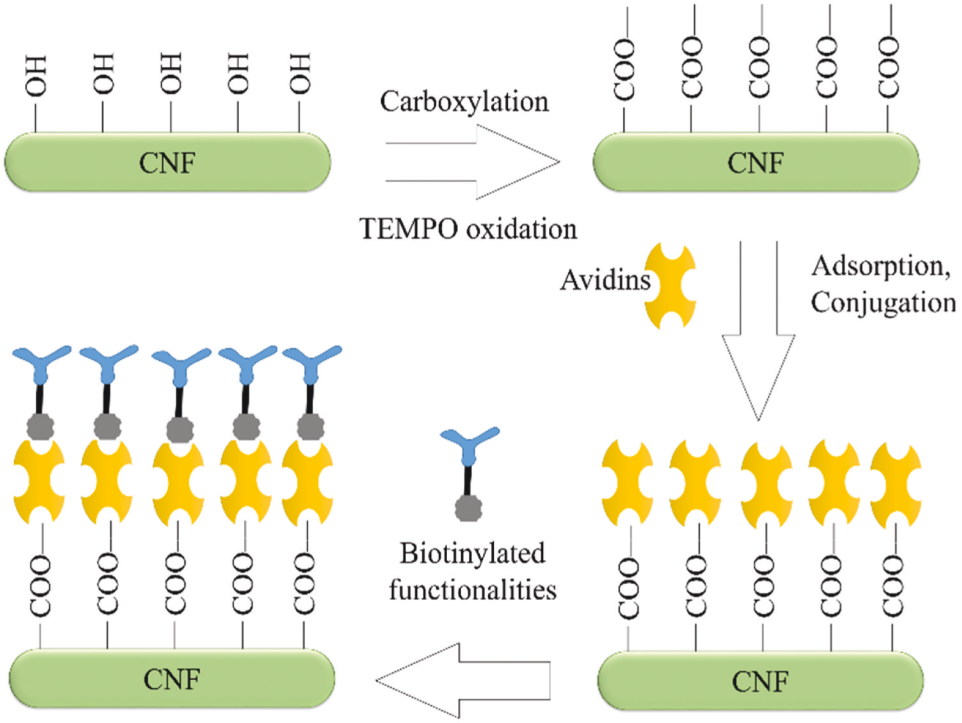

**Figure 9.** Schematic illustration for the preparation of antibody-functionalized cellulose surfaces via avidin – biotin interactions. Reproduced with the permission from (Orelma et al. 2012).

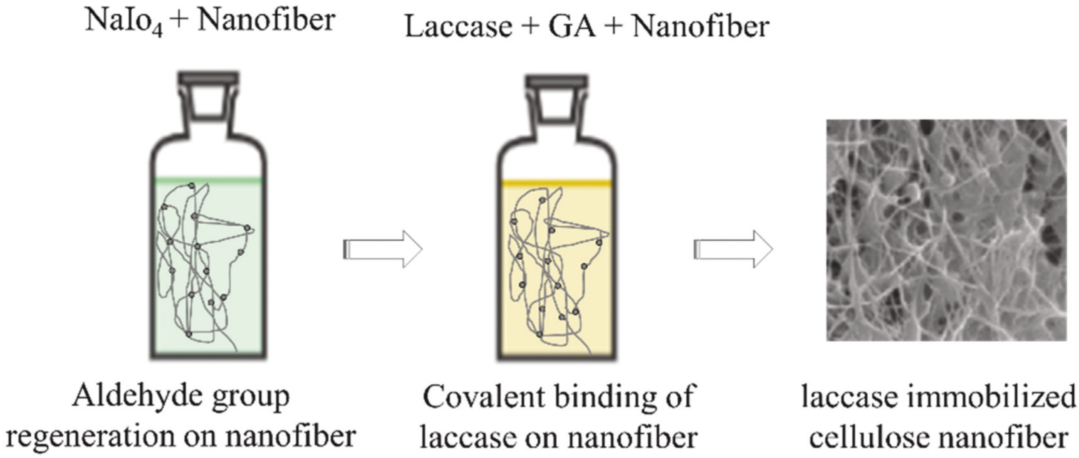

**Figure 10.** Approach of cellulose nanofiber laccase immobilization synthesis. Reproduced with the permission from (Sathishkumar et al. 2014).

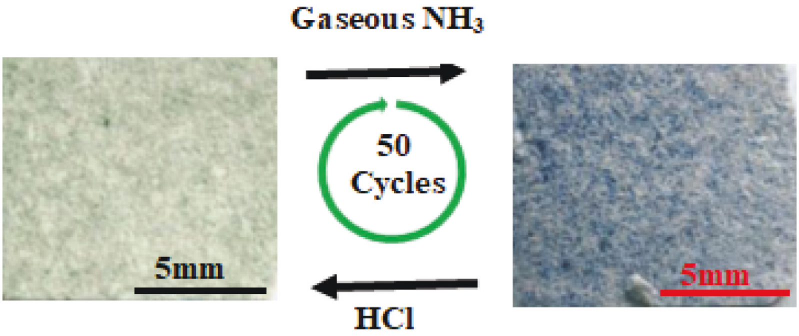

**Figure 11.** Color-changing behavior of CNF film coated by thin polyaniline films. Reproduced with the permission from (Gu and Huang 2013).

Another approach was also proposed by Davarayn and Kim. in developing an eco-friendly pH sensor via immobilizing a natural pigment onto the electrospun CNFs. In this study, natural pigment immobilization was carried out via adsorption and chemical crosslinking reaction by a bifunctional cross-linker reagent (hexamethylenediisocyanate), and different colors were observed for different pH values in the range of 1 – 14 (Devarayan and Kim 2015). Chemical remote detection via colorimetric approaches is another potential application introduced for CNFs. In this regard, Gu and Huang fabricated a sensitive and reversible colorimetric gaseous ammonia sensor by coating CNFs with thin-polyaniline films via *in-situ* polymerization of aniline molecules. The results revealed a quick color change from green to blue within 5 min at 30°C in the presence of gaseous or aqueous ammonia (Figure 11). Moreover, the recyclability in colorimetric sensing of CNFs sheet at least 50 times confirmed the possibility of a facile and effective route to improving the sensitivity of colorimetric gaseous ammonia detectors (Gu and Huang 2013).

## Transparent electric devices

Future, efficient, and sustainable transparent electric devices would require new materials characterized not only by high optical transparency, and electrical conductivity, but also by mechanical strength, thermal stability, and flexibility. Cellulose nanofibers thanks to their extraordinary mechanical and optical properties, high specific surface area, and low coefficient of thermal expansion have more recently been explored as emerging and advanced players in the fabrication of bio-inspired transparent devices (Pakharenko et al. 2021). One of the important drawbacks to using plastic-based transparent devices is their thermal dimensionally unstable properties. It is well addressed that CNFs are able to meet all of the requirements for such advanced and multifunctional applications especially high thermal stability (Biswas et al. 2019; Subir Kumar Biswas et al. 2019; Tanpichai et al. 2020). Biswas et al., have fabricated a novel nanocomposite with cellulose nanorod-encapsulated acrylic resin. They have reported that this nanocomposite with only ~10 wt % CNFs content possesses hierarchical nanostructure and very high thermomechanical, thermodimensional, and thermooptical stability for temperatures of ~150 – 180°C. It is also addressed that CNFs can be applied to generate a nanocomposite that possessed high mechanical properties even at an elevated temperature of 150°C and good electro-optical properties (12.4–15.6 Ω sq – 1 vs 84%), which were interestingly comparable to or even better than those available commercially (Biswas et al. 2019). Tanpichai et al., prepared CNFs-based nanocomposite which tough and optically transparent properties and is promising to be applied as an alternative to glass substrates in electronic applications such as flexible displays and solar cells (Tanpichai et al. 2020). Pakharenko et al., recently designed and fabricated thermo-stable cellulose nanofibers thin-films up to 190°C for 5 h of exposure and over 10 years equivalent in use the life span of a lighting device designed to achieve good durability (Pakharenko et al. 2021).

## Cellulose nanocomposites

### Cellulose nanocomposite preparation; opportunities and challenges

Cellulose nanocomposite study dates back to the mid-90s when Favier's research group investigated the application of monocrystals of cellulose with an aspect ratio of around 100 and an average diameter of 20 nm in improving mechanical properties of latex nanocomposites (Favier et al. 1995). Then after, many researchers apply their own perspectives to prepare cellulose nanocomposites using different techniques (Moon et al. 2011). The main challenge in attaining superior performance of nanocellulose as a reinforcing agent lies in achieving the homogeneous dispersion of nanofillers within the polymer matrix and an optimum matrix – filler interaction. However, the high hydrophilicity character of cellulose nanofillers results in the formation of aggregates due to van der Waals bonding, and this, in turn, leads to an inefficient dispersion of the cellulose nanofillers, weak interaction between the matrix and reinforcing agent, and limit the actual potential of mechanical reinforcement. Also, this phenomenon is magnified when the size of the particle decreases. Therefore, the research on the manufacturing process of cellulose nanocomposites aims at optimizing cellulose nanofiller dispersion through host polymer matrices through different manufacturing processes. In general, the application of pre-treatment of nanocelluloses prior to composite processes is an essential step in ensuring optimized nanocellulose dispersion through polymer matrices.

### Surface modification of cellulose nanomaterials

In nanocomposite materials, the surface characteristics of cellulose nanomaterials govern the interaction and interfacial adhesion between the nanofiller – polymer matrices. This in turn can alter the final properties of corresponding nanocomposites as the optimum nanofiber – polymer interaction is required for load transition from the host polymer matrices to nanofiller (Pan et al. 2018). The presence of numerous hydroxyl groups on the surface of cellulose nanomaterials allows the alteration of the surface of CNMs by imparting new characteristics while maintaining the original morphology of the nanocrystals (Lin and Dufresne 2014). In general, covalent or noncovalent coupling of hydrophobic moieties on the surface of cellulose nanoparticles is the most common approach in many cellulose nanofillers' surface modification treatments. The high reactivity of the hydroxyl groups to

**Table 1.** Different chemical modification approaches which have been employed for cellulose nanostructures (CNCs and CNFs) modification in nanocellulose-composite preparation.

| Surface modification treatment | Type of cellulose nanomaterials | Host polymer matrix | Reference |
|---|---|---|---|
| Salinization | CNFs | Polysulfone | (Zhang et al. 2018) |
| Polymer grafting | CNFs | Polycaprolactone | (Boujemaoui et al. 2017) |
| Alkylation | Microfibrilleted cellulose (MFC) | Low density polyethylene (LDPE) | (Lepetit et al. 2017) |
| Alkylation | Microcrystalline cellulose (MC) | High density polyethylene (HDPE) | (Sato et al. 2019) |
| Acetylation | CNCs | Poly (3-hydroxybutyrate-co -3-hydroxyhexanoate) (PHBH) | (Li et al. 2019) |
| Esterification | CNCs | Polylactide acid (PLA) | (Shojaeiarani, Bajwa, and Hartman 2019) |
| Esterification | CNCs | Epoxy | (Trinh and Mekonnen 2018) |
| Glyoxalization | Bacterial cellulose (BC) | PLA | (Quero et al. 2012) |
| Acetylation | CNFs | PLA | (Ying et al. 2018) |
| TEMPO | CNFs | Poly(N-isopropylacrylamide) (PNIPAm) | (Wei et al. 2016) |

form hydrogen bonds strongly influences the overall properties of nanocellulose such as the reactivity of the hydroxyl groups, hierarchical organization, crystallinity, and limited solubility in most solvents (Habibi, Lucia, and Rojas 2010). The strong hydrophilic character of cellulose nanocrystals due to the presence of free hydroxyl groups on the surface of CNCs restricts the application of different solvents processing mediums for solution blending (Nagalakshmaiah, El Kissi, and Dufresne 2016). In addition, the dispersion of CNFs into the hydrophobic polymer and water-insoluble nature is still a big challenge. Therefore, several techniques have been experimented with to decrease the affinity for moisture and improve the compatibility with a nonpolar polymer. The presence of hydroxyl groups on the surface of CNFs provides the opportunity for the application of different surface modification techniques to alter the hydrophilicity and improve the compatibility with different nonpolar polymer matrices. Much research has been devoted to moderating the structure and hydrophilicity of cellulose nanofibers using physical and chemical modifications. Table 1 summarizes some examples of chemical modification methods which have been employed for CNFs and CNCs in nanocomposite preparation.

In all aforementioned studies, the noticeable improvement in mechanical characteristics of nanocomposites was more likely attributed to the uniform dispersion of nanofillers through host polymer matrices. However, the major limiting factor in cellulose nanocrystals surface modification treatment is the tedious solvent exchange process and the use of organic solvents in these reactions. Recently, a green surface modification treatment of cellulose nanocrystals was reported by Espino et al. Benzoic acid was used as a grafting agent on the surface of CNCs and solvent media above its melting point (Espino-Pérez et al. 2014). Later, Shojaeiarani et al., showed that such environmentally friendly surface modification treatments have the potential to be applied for nanocomposite manufacturing (Shojaeiarani, Bajwa, and Stark 2018).

## *Solvent casting*

Besides chemical surface modification treatment, numerous studies have focused on the application of mechanical pre-processing techniques to maximize the dispersion quality of nanofillers in the host polymer matrices (Chanda and Bajwa 2021). Solvent casting or wet blending is an old-time thin film-making technology exploited by numerous researchers to provide optimum control of film thickness. In solvent casting, the host polymer is first dissolved in a volatile solvent and followed by the incorporation of the nanofillers. In some works, high power sonication is used to ensure the dispersion of nanofillers in polymer solution (Beuguel et al. 2018; Oun and Rhim 2016; Xiao et al. 2015) and improve the interaction between polymer and nanofillers. It is worthy to say that the interactions between the cellulose nanofillers and solvent or polymer solution can be improved by the reduction of the van der Waals force between nanofillers, therefore assisting the polymer chains to diffuse into the seats among nanofillers (Atif and Inam 2016). After mixing the polymer solution and nanofiller suspension, a solid nanocomposite film can be obtained by solvent evaporation. However, the slow solvent evaporation rate in the solvent casting method favors the formation of cellulose nanofiller aggregates and prevents the commercial application of solvent casting at the industry level. In addition, the dried solvent-cast thin films can contain organic compounds like residual solvents, which resulted in a reduction in nanocomposite characteristics. Therefore, along with ultra-sonication treatment, supplementary processes, such as magnetic (Mao et al. 2018) and electric fields (Frka-Petesic et al. 2017) surfactant (Kaboorani and Riedl 2015), are applied to cellulose nanocrystals suspension to reduce the negative aspect of slow solvent evaporation rate in solvent casting method. The solvent casting technique has been extensively employed to fabricate the highly concentrated cellulose nanocrystals master-batch as a preprocessing technique in nanocomposite manufacturing. In general, the master-batch approach is a relatively new way to prohibit CNCs aggregation during the drying stage before extrusion. The two-stage process includes preparing a high-CNCs content thin-film masterbatch, then diluting the masterbatch via mixing polymer host with the masterbatch, and preparing the nanocomposite with desired CNCs concentration in the extruder (Sapkota et al. 2017; Shojaeiarani, Bajwa, and Hartman 2019). In this regard, Pracella's research group proposed a novel method for preparing PLA-reinforced CNC nanocomposites using glycidyl methacrylate radical grafting and CNCs master-batch in a PVA matrix (Pracella, Haque, and Puglia 2014). They confirmed a more uniform dispersion of CNC in polymer matrix after premixing in PVA matrix. In a similar work, Mariano et al., employed a master-batch approach followed by twin-screw extrusion and injection molding to prepare PLA nanocomposite reinforced by CNCs. The noticeable improvement in thermal stability and CNCs dispersion quality in polymer matrix during extrusion was attributed to the application of the master-batch approach (Mariano et al. 2017).

### Continuous melt extrusion process

Melt extrusion or melt blending is an ideal processing method for preparing polymer-based nanocomposites of thermoplastics and the elastomeric polymeric matrix (Azouz et al. 2012). In this method, the polymer is heated above its glass transition temperature (*Tg*) and forced through an orifice in a continuous process, where the polymer pellets are continuously fed into the hopper, melted, and mixed (Figure 12). Continuous processes are favorite for scaling up as it has better mixing and venting performance in comparison with batch extrusion. Melt extrusion is highly automated, efficient, and environmentally friendly as it is a solvent-free method, however, this method is limited for sensitive polymers due to thermal degradation as a result of high

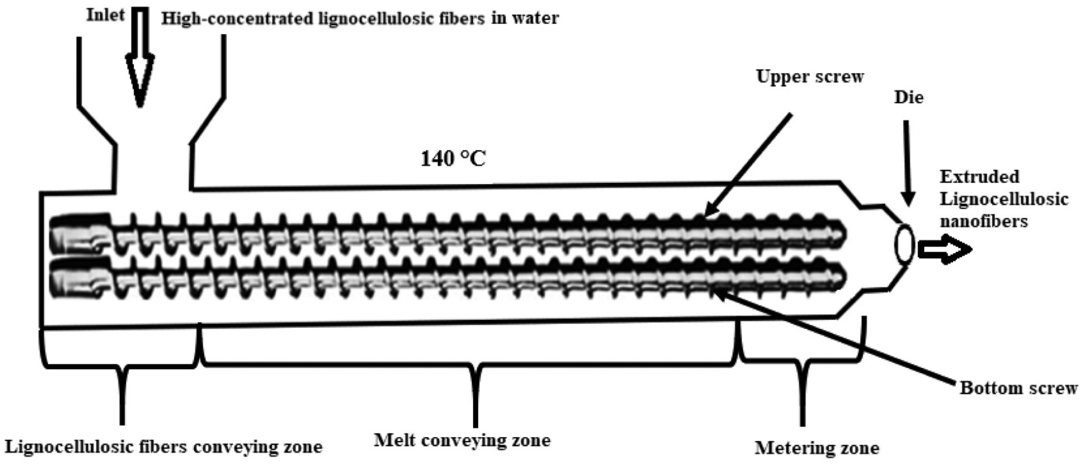

Figure 12. Schematic illustration of a typical twin-screw extruder with its different sections.

temperature (Wang et al. 2020). Among different melt extruders, the screw extruders are the most common type in the polymer industry as they continuously convert polymer pellets into a finished form such as a filament or extrudates. Melt extrusion can be conducted by a twin or single screw extruder. Intermeshing twin-screw extruder is often used to process highly viscous materials and for the removal of large extents of volatile substances, while simple screw extruders have better performance in mixing materials (Patil, Tiwari, and Repka 2016)

Menezes et al. reported an attempt in preparing LDPE reinforced by cellulose whiskers using high shear melt extrusion technique. They employed a twin-screw extruder to mix LDPE and cellulose whiskers and push them through die (Junior de Menezes et al. 2009). Melt extrusion has attracted significant attention in preparing cellulose nanocomposites as several researchers used it not only for polymer-nanofiller mixing but also for diluting masterbatches and providing the nanocomposites with desired nanofiller contents (Abdallah et al. 2019; Karkhanis et al. 2018).

### *Batch mixing process*

The batch mixing process is a favorite technique to fabricate nanocomposite materials on small scales (Ojogbo, Tzoganakis, and Mekonnen 2021). Batch mixers are mainly consisting of the two-roll mill and internal mixers capable of melting and mixing polymers. In these processes, polymers have high-chain flexibility through the input of thermal energy and are mixed with the nanofillers mechanically (Kim et al. 2007). The long processing time is one of the most important drawbacks limiting the application of this method on a large scale since a long processing time would result in discoloration and degradation of the polymer or the cellulose (Gu et al. 2018). It was also reported that the shear forces present in a batch mixer could not prevent the agglomeration of fibers during nanocomposite manufacturing. However, A number of cellulose nanocomposite researches have been conducted using a batch mixer to distribute cellulose nanofiller in the melt, and an extruder to prepare nanocomposite(Abdallah et al. 2019; Sapkota et al. 2017; Sojoudiasli, Heuzey, and Carreau 2018). Furthermore, the addition of some chemical grafting such as polyethylene glycol inhibited the thermal degradation and discoloration of cellulose nanocomposites and improve the thermal stability of nanocomposites (Lin and Dufresne 2014). Rogovina et al. prepared a biodegradable composite of polylactide with ethyl cellulose and chitosan plasticized by low-molecular poly(ethylene glycol) (Rogovina et al. 2016). They mixed the desired amount of cellulose and chitosan in a Brabender mixer at 160° C for 10 min. Then, they prepared composite thin films with the thickness of 0.18–0.25 mm at 160° C for 10 min using a hot press. In a different work, Lima's research group employed a batch mixer and mini-twin screw extruder to produce a conductive cellulose acetate (CA)-carbon nanotube nanocomposite with cellulose acetate as host polymer (Delgado-Lima, Paiva, and Machado 2017). Roll-milling is another batch processing that applies medium and high shear forces in nanocomposite manufacturing mainly used for dispersing cellulose nanocrystals in thermoset resin in a controlled temperature situation to prevent premature resin curing (Puglia and Kenny 2018). In this regard, Shin et al., employed three-roll milling and probe sonication combination to prepare CNCs reinforced epoxy (Shin et al. 2017).

### *Pickering emulsion approach*

Pickering emulsions are recently introduced as a novel and advanced approach to fabricate the nanocellulose-based composites (Fujisawa, Togawa, and Kuroda 2017). Pickering emulsions are emulsions stabilized by solid particles. Compared to surfactant molecules, the solid particles more strongly adsorb at liquid/liquid interfaces. Therefore, Pickering emulsions generally show better stability than emulsions stabilized by surfactants (Fujisawa 2021). A CNF-stabilized monomer-in water emulsion was used as a starting point to design a CNF/polymer composite structure (Fujisawa, Togawa, and Kuroda 2017). Several research have been conducted on CNFs and LCNFs-based nanocomposite preparation through Pickering emulsions (Biswas et al. 2017, 2019; Sato et al. 2019; Yan et al. 2016). Furthermore, the synthesis of core-shell microparticles using Pickering emulsion has been reported (Fujisawa et al. 2019). There would be an increasing interest to fabricate a scaffold for biomaterial applications such as cartilage, bone, nerve, and cardiovascular tissue regeneration with this approach thanks to less toxicity and no chemistry use (Tanpichai 2022).

## *Advanced tools to characterize the nanocellulose-based composites*

In order to optimize the performance characteristics of polymers reinforced by nanocellulose, rapid screening and characterization of nanocomposites using a wide array of sophisticated techniques have been extensively conducted. The overall performance characteristics of cellulose nanocomposites are mainly evaluated using a combination of different techniques including morphology, thermal, mechanical, and rheological testing depending on the final applications of proposed nanocomposites. Specifically, the dispersion quality and nanofiller dimension into host polymer matrices play a key role in different characteristics of the nanocomposite. Different microscopy techniques have been applied to extensively evaluate the structure, size, and morphology of nanofillers in the host polymer matrices. In this regard, Optical microscopy (OM), Transmission electron microscopy (TEM), Scanning electron microscopy (SEM), Atomic force microscopy (AFM), and Raman spectroscopy with different resolutions have been conducted to study the nanocellulose in the polymer matrix (Bulota et al. 2012; Tanpichai, Sampson, and Eichhorn 2012). For all different testing methods, the preparation process is an essential step, as cellulose nanocomposites are non-conductive, soft, and hydrophilic materials (Ramos and Matouš 2018).

## *Transmission electron microscopy (TEM)*

Transmission electron microscopy (TEM) is an analytical method, that transmits an electron beam through an ultrathin specimen with a thickness lower than 1 μm or a sample suspension on a grid. In the case of CNCs imaging, the suspension is deposited on a thin circular metallic grid with typical meshes in the order of tens of micrometers. There are different types of grids such as copper, molybdenum, gold, or platinum. The grid selection depends on different factors such as CNCs suspension pH. For relatively high or low pH, gold, or nickel grids can be used to avoid degradation and the formation of artifactual crystallites or dendrites upon drying. For the supporting films, carbon film with a typical thickness of 5–20 nm is the most commonly used material. Formvar is a coating material mainly used to coat copper grids, however, it is less electron-transparent than carbon and can drift when heated by the electron beam at high magnification (Gontard et al. 2014).

Focused ion beam microscopes (FIBs) are used to prepare TEM specimens in laboratories, however, the non-conductive character of nanocellulose, low density, and high sensitivity against electron beam damage make this process challenging. In this regard, selective staining agents can be used to create an electron-dense outline for individual CNCs, however, special care must be taken to prevent the layer of dye from overshadowing the contrast of nanofibers. For instance, the presence of too much background stain might negatively influence the TEM micrographs. In general, sample staining is considered in TEM imaging to absorb the beam electrons or scatter part of the electron beam and improve the contrast. Among different negatively charged stains, uranyl acetate solution (2% w/v) is the most common stain used for preparing CNCs for TEM imaging (Kaushik 2015). Figure 13 demonstrates the TEM images of the CNCs and CNFs from different raw materials.

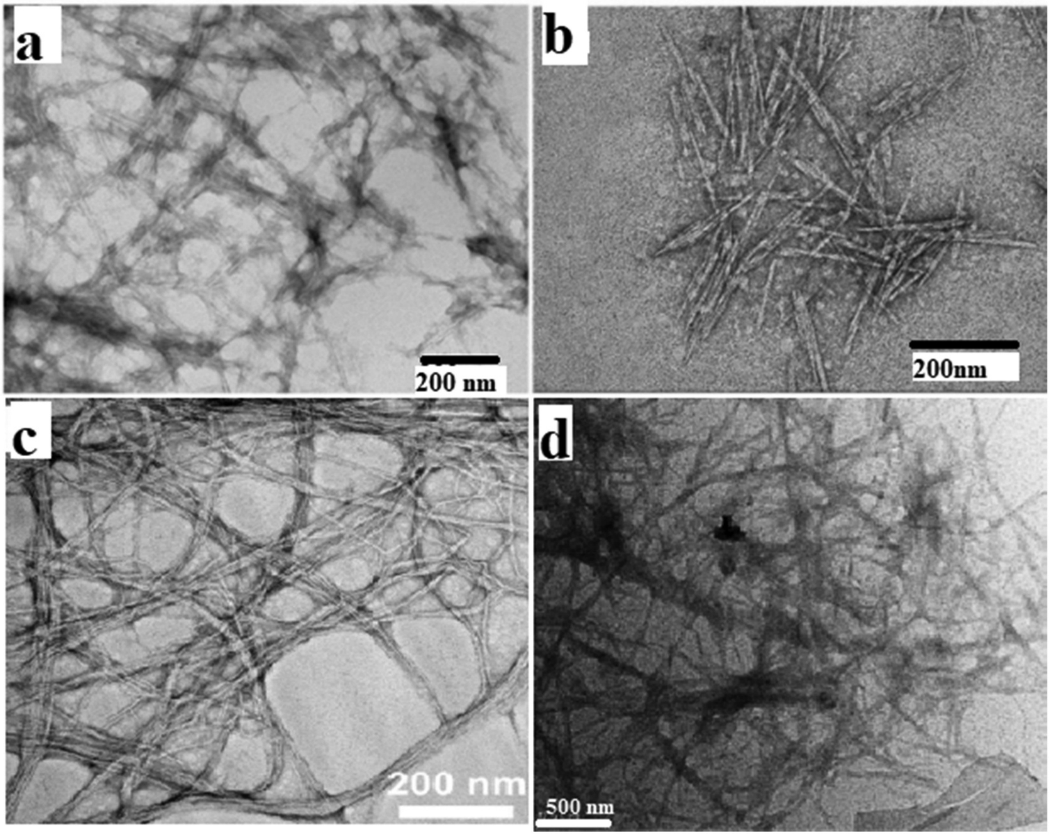

**Figure 13.** TEM images of CNCs from (a) Jujube Cores at 30,000 magnifications (Wang et al. 2018) (b) from Jujube Cores at 40,000 magnifications (Wang et al. 2018), and CNFs from (c) bleached softwood sulfate pulp through microfluidization (Nissilä et al. 2021) (d) enzymatic pretreated industrial waste sludge (Gallo Stampino et al. 2021).

## *Scanning electron microscopy (SEM)*

A scanning electron microscope (SEM) is one of the most adaptable instruments available for the surface topography assessment of the micro and nanostructure morphology of cellulose-based nanocomposites, discriminating objects on a nano-scale. The SEM micrographs are mainly used to represent the nanocomposite characteristics as a function of nanofiber size, shape, orientation, and the matrix-nanofiber interaction (Dikin et al. 2022). It includes finely collimated electron beams that swept across the surface of the specimen and illustrate the surface topography. FESEM image of mechanical entanglement of CNFs in packaging paper at different magnifications (Figure 14).

When the electron beam hits the surface of the sample, it penetrates the sample to a depth of a few microns, depending on the accelerating voltage and the density of the sample. Many signals, like secondary electrons and X-rays, are produced because of this interaction inside the sample (Figure 14). The resolution of SEM images is generally depending on the electron beam spot size, which is smaller at higher acceleration voltage (AV) (Table. 2).

For sample preparation, the surface of nanocomposite samples must be electrically conductive, and electrically grounded to a sample holder to avoid the electrostatic charging at the surface of the sample during electron irradiation. The SEM imaging of cellulose nanocomposites is challenging due to the non-conductive nature of CNCs which results in nanocomposite charging, scanning faults, and image

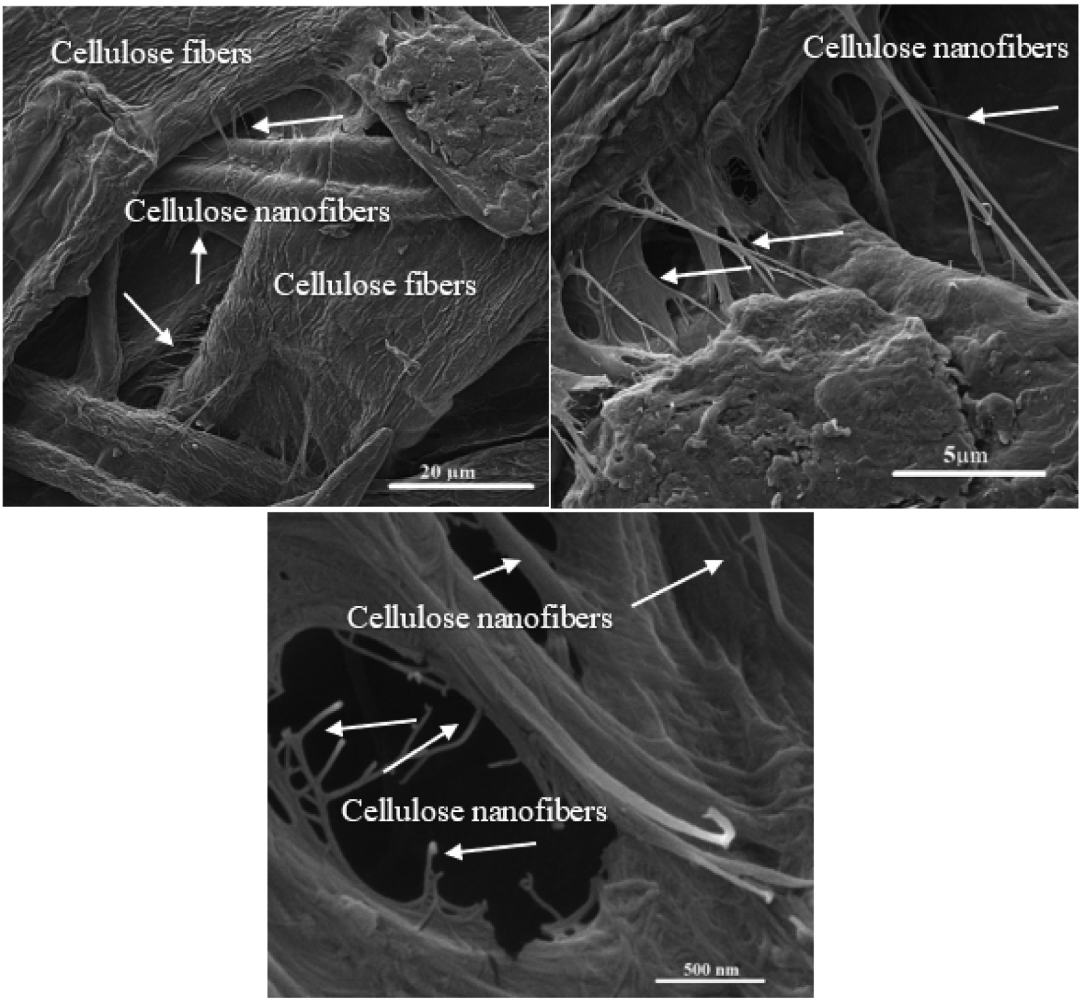

**Figure 14.** FESEM image of mechanical entanglement of CNFs in packaging paper at different magnifications (a) 2.50 kx (b) 10 kx (c) 75.0 kx. The white arrows indicate the additional cellulose nanofibril structures which advance the binding between each other and fibers.

**Table 2.** Advantages and disadvantages of high or low accelerating voltage (Larissa Do Amaral Montanheiro et al. 2019; Mahmud et al. 2019; Mondragon et al. 2018; Pereira Novo, Curvelo, and Carvalho 2018).

| Accelerating Voltage | Advantages | Disadvantages |
|---|---|---|
| High | High Resolution | Unclear surface structures<br>More edge effects<br>More charge-up<br>More damage |
| Low | Clear surface structures<br>Less damage<br>Less charge-up<br>Less edge effect | Low Resolution |

artifact. Therefore, the sample preparation is an important step, and nanocomposites are usually coated with an ultra-thin layer of conductive materials such as platinum, gold, or carbon using a vacuum sputtering machine (Zhan et al. 2017). In the case of cellulose nanocrystals, SEM micrographs are mainly used to evaluate the nanocomposite surface morphology(Larissa Do Amaral

Montanheiro et al. 2019; Wu et al. 2018) and polymer and CNCs interaction (Pereira Novo, Curvelo, and Carvalho 2018). The SEM is also widely applied to evaluate CNCs dispersion in a host polymer matrix on impact or tensile (Mondragon et al. 2018) and address the fracture surface of the prepared nanocomposites (Mahmud et al. 2019)

## *Atomic force microscopy (AFM)*

Atomic force microscopy (AFM) is a powerful mechanic-optical instrument creating three-dimensional images at angstrom and nano-scale from the surface of the sample. AFM conducts surface scanning using the probing stick which is a micro-sized cantilever with a tip at the free end. A piezo controller swings the probe over the surface in three dimensions with nanometric precision. The accurate position of the tip over the sample surface and how strongly the tip slides or taps over the sample surface is controlled by a detector. The AFM analysis has several advantages over SEM and TEM since no special preparation is required for the sample. Furthermore, AFM imaging can be conducted under atmospheric pressure and detailed features such as surface forces (adhesion, friction, electrostatic, van der Waals, etc.) can be observed from AFM images (Dufrêne 2002). AFM has been widely used for the study of cellulose nanocomposites as the surface of nanocomposites presents in-homogeneities on the nanoscale (Figure 15).

Properties such as adhesive energy maps (Younas et al. 2019), transverse elasticity, surface topography (Mitrovic et al. 2018), and axial elasticity (Wu, Moon, and Martini 2014) have been widely studied using AFM. Different researchers also evaluated the dispersion of CNCs in polymer matrices using AFM images (Dhar et al. 2016). There are different AFM modes for measuring the elastic properties of nanocomposite components such as the force-displacement method (Pettersson et al. 2017), force modulation microscopy (Pakzad, Simonsen, and Yassar 2012), and contact resonance AFM (Wagner, Moon, and Raman 2016). Alam's research group employed AFM to compare the physical dimensions of single nanocellulose blended with conducting polymer in vertical and lateral directions. They validated AFM results using DLS and MASLS observations (Alam et al. 2019). In a different approach, AFM images were obtained to evaluate the microstructure of samples and microphase separation of hard and soft segments in polyurethane-CNC nanocomposites. In which, brighter regions represented hard segments and darker areas related to soft segments (Khadivi et al. 2019) (Figure 16).

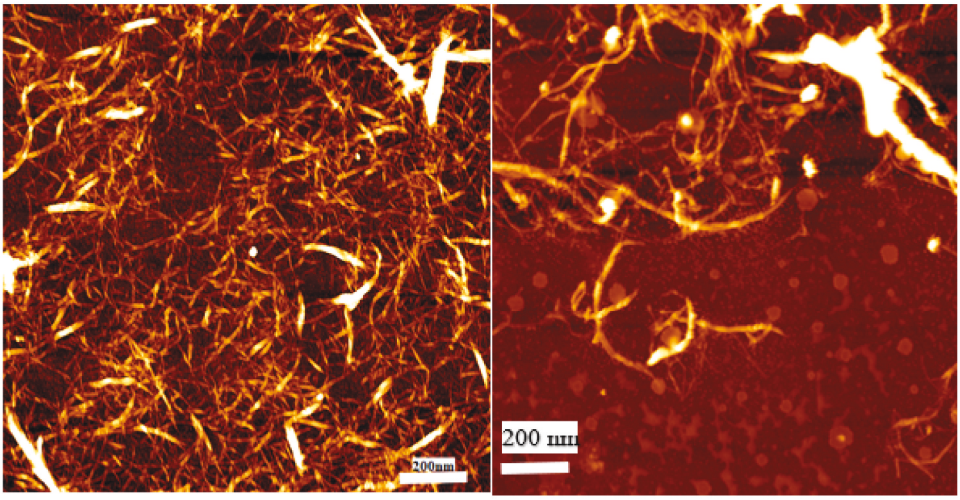

**Figure 15.** AFM images of (a) CNCs (b) CNFs extracted from giant reed (*arundo donax*).

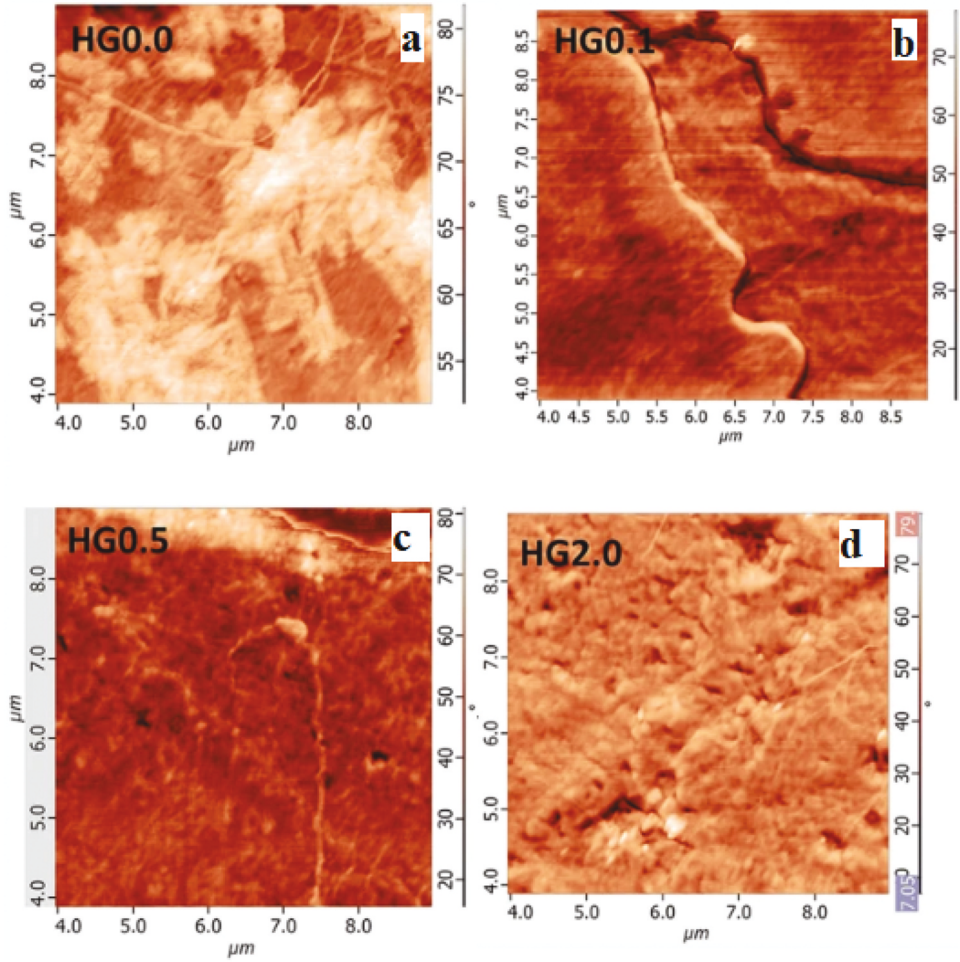

Figure 16. Different color regions in AFM images represent microphase separation in polyurethane-CNC nanocomposites: HG means (Hexamethylene diisocyanate (HDI) and PEG-based PU, and the number after HG represents the amount (wt.%) of CNCs: (a) Neat PU (b) 0.1 CNC/PU (c) 0.5 CNC/PU (d) 2 CNC/PU (Khadivi et al. 2019).

## *Raman spectroscopy*

Raman spectroscopy is a powerful technique to investigate the molecular deformation of different bio-based fibers, including cellulose-based composites (Bulota et al. 2012; Tanpichai, Sampson, and Eichhorn 2012). This technique actually relies on the measurement of a change in the position of a Raman band on the application of external tensile deformation (Eichhorn and Young 2001). Recently, this well-known technique has been used to study the stress-transfer process in microfibrillated cellulose-based composites (Tanpichai, Sampson, and Eichhorn 2014), bacterial cellulose-based, and CNFs-based composites (Bulota et al. 2012) through measuring a shift in a Raman peak initially located at ~1095 cm$^{-1}$, attributed to C–O–C stretching of the nanocellulose fractions into the host polymer matrix. It is well shown that stress transfer in the nanocellulose-reinforced composite greater than that of a pure network of fibers, indicating a good fibre – matrix bond (Bulota et al. 2012; Tanpichai, Sampson, and Eichhorn 2012, 2014). It is worth mentioning that, this technique is well addressed as a powerful one to study the orientation of nanocellulose fibril into the hot-polymer matrices by rotating the samples by 5°

angular increments from 0 up to 360°. The intensity of the Raman peak located at ~1095 cm – 1 was continuously recorded at each rotation increment (Bulota et al. 2012).

## Conclusions and future outlook

Cellulose as an abundant bio-inspired polymer possesses engrossing properties such as excellent biocompatibility, low cost, low density, high intrinsic strength, and good mechanical performance. Cellulose nanomaterials are also well recognized as extraordinary nano-biomaterials that have received tremendous consideration to be applied in high-performance and multifunctional application fields. The inimitable characteristics of CNMs are due to their unique structural, physicochemical, mechanical, and biological properties, and availability to be modified by various chemical reactions. This review aims to provide an overview of several aspects of cellulose nanocomposites fabrication from preparation, classification, surface modification, nanocomposite fabrication, characterization, and their potential applications. Then, surface modifications toward CNMs have been elaborated as an applicable strategy to overcome their intrinsic limitations to fabricate high-performance nanobiocomposites. The review also brings an insight into sophisticated and advanced characterization tools of the nanocellulose-based composites. It has also been shown that the CNMs especially CNCs may improve the crystallinity and the rigidity of the host polymer matrices. In short, this review well addresses the advanced emerging applications of CNMs *i.e.* biosensors, electromagnetic shielding, sustainable packaging, and bio-medical field that are able to profit from the unique properties of CNMs and proposes the future prospects for their commercialization. Overall, despite all the research done yet, the authors believe that such a comprehensive study is still required in this era.

## Highlights

- Current trends and prospects in cellulose nanomaterials synthesis are reviewed.
- Cellulose-based nanocomposites can be applied in a broad range of potential applications.
- High-performance, multifunctional, and emerging potential applications of nanocellulose-based composites are addressed.
- Sophisticated fabrication and characterization techniques are promising to advance the specific cellulose nanocomposite functions.

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We confirm that this research meets ethical guidelines and adheres to the legal requirements of the study country. The research does not involve any human or animal welfare-related issues.

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