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## Application of nanocellulose composites in the environmental engineering as a catalyst, flocculants, and energy storages: A review

Mario Pagliaro <sup>a</sup>, Rosaria Ciriminna <sup>a</sup>, Mohammad Yusuf <sup>b</sup>, Sara Eskandarinezhad <sup>c\*</sup>, Irshad Ahmad Wani <sup>d</sup>,

Mina Ghahremani <sup>e</sup>, Zahra Rezaei Nezhad <sup>f</sup>

<sup>a</sup> Istituto per lo Studio dei Materiali Nanostrutturati, CNR, via U. La Malfa 153, 90146 Palermo, Italy

<sup>b</sup> Department of Chemical Engineering, Universiti Teknologi PETRONAS, Bandar Seri Iskandar, 32610, Malaysia

<sup>c</sup> Department of Mining and Metallurgy, Yazd University, Yazd, Iran

<sup>d</sup> Postgraduate Department of Chemistry, Govt. Degree College Bhadarwah, University of Jammu, Jammu and Kashmir, India, 182222

<sup>e</sup> Faculty of Chemistry, Institute for Advanced Studies in Basic Sciences (IASBS), Gava Zang, PO Box 45195-1159, Zanjan, Iran

<sup>f</sup> Department of Physical Chemistry, Faculty of Chemistry, Shahid Beheshti University, Tehran, Iran

### ABSTRACT

Nanocellulose, the most promising bionanomaterial, is obtained either from the degradation of natural polymers or by the activity of bacteria and microorganisms. These biomaterials present various advantages, including full recyclability, biodegradability, and lack of harmful effects on the human body and environment. Furthermore, nanocelluloses are candidates to fabricate thin transparent layers, fibers, hydrogels, and aerogels due to their remarkable optical, thermal, and mechanical behaviors, including high crystallinity, Young's modulus, and porosity content. These exceptional properties present the superb potential of these materials for the device of environmentally engineered tenable products. This paper presents an outline of the contemporary nanocellulose research works as well as details and information on the nanocellulose materials, especially the synthesis process of composites, along with the areas in which these materials can be utilized, such as energy, flocculant, pollution sensors, and catalysts, to respond to the rising requests of these materials.

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### 1. Introduction

In a report named “Global Nanocellulose Fiber Market Report” pub-

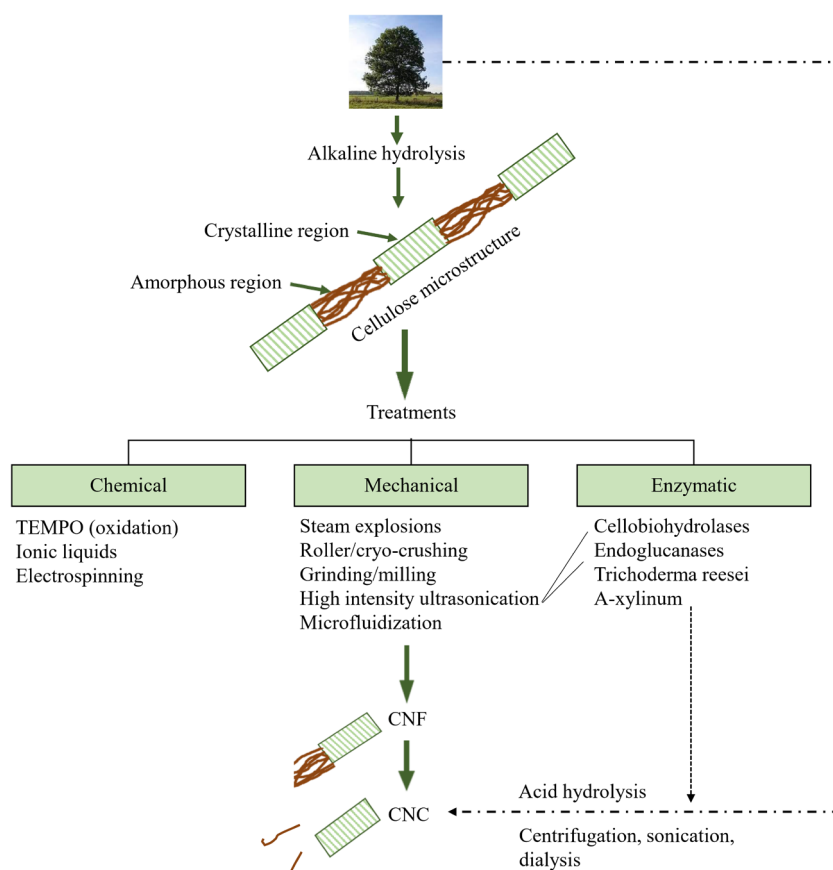
lished by Research Insights in 2020, it has been anticipated that the price of nanocellulose on the market will have reached \$2.712 billion by 2025, with an expansion rate of 18.80% per year through 2018 and 2025 [1]. Nevertheless, achieving this significant economic rise of nanocellulose

\* Corresponding author: Sara Eskandarinezhad; E-mail: [s.eskandari.nezhad@gmail.com](mailto:s.eskandari.nezhad@gmail.com)

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**Fig. 1.** The schematic of the CNFs from wood fibers using chemical and mechanical approaches.

materials largely depends on the production (e.g., the evolution of techniques, considering environmental matters), commerce and advertising (e.g., public approval, market penetration, cost managing), and human health matters, for example when applied in nutriment and biomedical industries) [2, 3]. At this time, only a handful number of companies have effectively taken nanocellulose materials and their valuable derivatives on the market. Nanocellulose materials are applied in several areas of our life due to their superior optical, mechanical, and thermal behaviors and characteristics. Despite that, to produce these materials on a commercial scale, their technical issues and high costs are still a challenge needing to be tackled. Regarding power supply and environmental aspects, nanocellulose is a source of recoverable energy, which is plentiful in terms of source variation and availability in nature. The advent of nanocellulose-based materials can save energy, solve environmental issues, and foster economic advances [4, 5].

During the past years, nanocellulose has exhibited convenient performance in a variety of applications such as paper production [6], the treatment of water [7]; and sewage [8], biomedicine technology [9, 10], and energy extraction [11]. Ranby et al. [12]; and Turbak et al. [13], first mentioned different types of nanocellulose in their articles to discuss scientific theoretical concepts and report new cellulose products, namely cellulose microfibril (CMF), cellulose nanocrystal (CNC), and micro fibrillated cellulose (MFC). Decades after that, active companies are currently producing nanocellulose with a range of size distribution and structures of nanoparticles [14].

The major content of the usual paper is cellulose, which has much larger diameters than nanocellulose (normally <10 nm) that presents unique properties, including great mechanical strength, high surface area, and limited scattering of the visible light. Both the decomposition of the cellulose pulp of plants and bacterial activity have been used to extract nanocellulose. These materials are employed for fabricating thin

translucent layers, fibers, hydrogels, and aerogels, exhibiting superior optical, thermal, and mechanical behaviors. These substrates act as an appropriate matrix or conveyor for inorganic nanoparticles, which results in the production of nanocomposites. Additionally, cellulose-based nanocomposites are easily attained through the direct combination of highly hydrophilic or hydrophobic nanocellulose derivatives with organic and inorganic substances. In principle, the abovementioned properties coupled to full biodegradability, recyclability, and lack of toxicity make these composites appropriate for application in a vast array of industrial fields [15, 16].

From the viewpoint of an environmentalist, nanocomposites based on nanocellulose, are promising materials for applications such as filtration of potable water [17], catalytic decomposition of organic-based contaminants [18, 19], spilled oil absorbents of water [20], and observing aquatic pathogens, and organic-based pollutants [21], and advanced energy conversion appliances [22]. In this review, the research scope focuses on the nanocellulose, chiefly nanocellulose-based nanocomposite, their synthesis procedures and their application in environmental-related science and engineering. In addition, the increasing and broadening demands to these materials and perhaps guide environmentalists and engineers interested in identifying new materials for handling current environmental issues is discussed in this review.

## 2. Nanocellulose

As nanotechnology has developed, nanocellulose-based materials have increasingly attracted the attention of scientists and scholars. Nanocellulose is a term used to explain nanomaterials based on cellulose with a minimum of one dimension, length, width, or breadth on the nanometric scale [23]. Nanocellulose is identical to the smallest constitutional component of the cellulose-based biomass of different organisms [24,

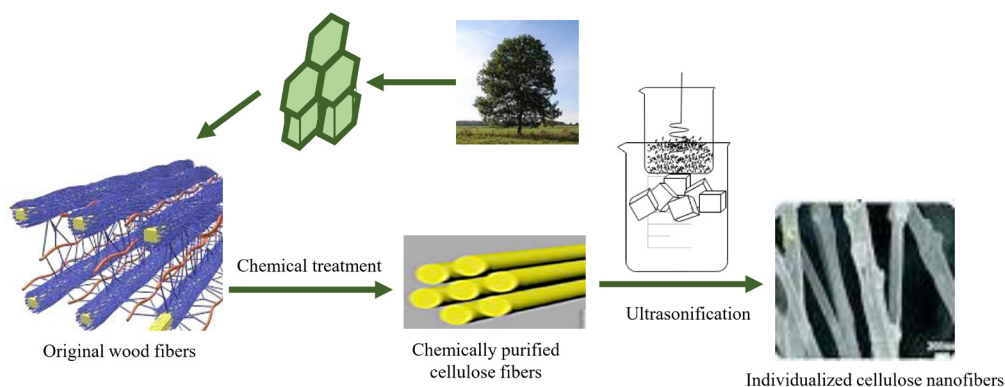


Fig. 2. Conventional methods used for obtaining CNC particles.

25]. The structure of nanocellulose is rod-shaped and rigid, with rod dimensions varying from 1 to 100 nm and 10 to 100 nm in diameter and length, respectively. Due to the abundance and bio-based nature of these materials, they are obtainable from several natural sources such as marine invertebrate animals, plant biomass, and algae. In addition, nanocellulose can be fabricated using some *Acetobacter* types of cellulose bacterial by the bio-based synthesis method [26]. The chemistry of nanocellulose is made up of a biopolymer in which d-glucoanhydro-pyranose units are repeated and linked by an ester bond ( $\beta 1 \rightarrow 4$ ) [27].

Similar to cellulose, nanocellulose presents an intrinsic chemical structure with ample hydroxyl groups and a specific quantity of aldehyde together with carboxyl groups to provide superior performance. Nanocellulose exhibits considerable properties, such as large surface area, active functional groups, high mechanical strength, and high crystallinity, due to its nanostructure, providing a large variety of applications in different fields [5, 28].

Conventionally, nanocellulose is obtained by two top-down and bottom-up technique, and the former involves mechanical, chemical, and enzymatic treatment of lignocellulosic fibers. For instance, agricultural biomass, including cotton fibers, ramie, hemp, and wood using a highly intensified ultrasonication approach into their respective fundamental nanofibrils with diameters inferior to 6 nm [29, 30]. Brown (1886) [31]; stated that a bottom-up method using fermentation of low molecular weight sugars and the *Acetobacter* types of bacterial cellulose could be employed to synthesize the fibers of nanocellulose or microbial cellulose.

These techniques, either used individually or successively, could obtain the desired structure and morphology [32]. The nanocellulosic materials are classified into three main subcategories based on their isolation methods. These categories include (1) cellulose nanocrystals (CNCs), also known as nanocrystalline celluloses (NCCs), nanocellulose whiskers (CNWs), and rod-like cellulose microcrystals, normally prepared from lignocellulosic materials and acid hydrolysis for removing non-crystalline cellulose from the cellulose nanocrystals with a rod structure (2). Nanofibers of cellulose (CNFs/NFCs), also named microfibrillated cellulose (MFC), nanofibrils, and microfibrils, are produced through mechanical methods or accompanied by enzymatic and chemical pretreatment methods for defibrillation of cellulose fibers. (3) Bacterial cellulose (BC), also known as bacterial nanocellulose (BNC), microbial cellulose, and biocellulose are commonly synthesized from various types of the *Acetobacteraceae* (Fig. 1) [33, 34].

Similar to other nanoparticles, such as carbon nanotubes (CNTs), nano-clay, and carbon black, various techniques have been used to characterize nanocellulose and its suspensions to study its surface chemistry, thermal stability, storage modulus, fiber length and width, fiber aspect ratio, complex viscosity, viscosity, aggregation degree, and crystallinity before its applications in different areas. The most commonplace

methods widely applied for the investigation and characterization of the behavior and characteristics of synthesized nanocellulose are X-ray photoelectron spectroscopy, differential scanning calorimetry, dynamic light scattering, dynamic mechanical analysis (DMA), zeta potential, scanning transmission electron microscopy (STEM), atomic force microscopy (AFM), X-ray powder diffraction (XRD), Fourier transforms infrared spectroscopy (FTIR), transmission electron microscopy (TEM), scanning electron microscopy (SEM), and thermogravimetric analysis (TGA) [35, 36].

### 2.1. Cellulose nanofibers (CNF)

Cellulose nanofiber (CNF), also called nanofibrous cellulose, nanofibrillar cellulose, and nanofibrillated cellulose (NFC), is characterized by its distinctive structure made up of a bundle of stretched cellulose chain molecules with cellulose nanofibrils together with repeating crystalline and amorphous regions [37–39]. NFC is composed of a mass of stretched nanofibers of cellulose. The cellulose chains possess a large surface area and are twisted and flexible. NFCs are different from CNCs in that they are composed of many amorphous domains and soft, long chains with widths and lengths often to a few hundred nanometers and several micrometers, respectively [40, 41]. Before converting the fibers to CNF, a vigorous mechanical disintegration should be exerted to the cell wall of the plant because of the sophisticated cellulose fiber structure. The fiber diameter is generally from 10 to 100 nm, dependent on the disintegration force [37, 38].

Various researches have focused on the CNF extraction from cellulose-based sources, including rice straw, oil palm biomass, flax, wood pulp, bamboo, cotton, hemp, and kenaf [42, 43]. The most important approaches for deriving CNFs from cellulose fibers are mechanical treatment such as milling, grinding, homogenization, chemical treatment, such as TEMPO oxidation, and a combined process of both chemical and mechanical treatments [44, 45]. Mechanical treatments are primarily used among all the treatment procedures. Figure 1 shows the cellulose nanofibers from wood fibers using chemical and mechanical approaches. In general, nanofibrillated cellulose defibrillation requires intense mechanical treatment processes such as grinding, microfluidization, cryo crushing, high-pressure homogenization, high-intensity ultrasonication, and a combination of two or three treatment processes [27, 42].

Unlike biological methods, including enzymatic treatments, chemical treatment is based on alkali treatments [46, 47]. Many researchers have applied the integration of these approaches to achieve the desired product. Similar to CNCs, the basic properties of NFCs change according to the source of raw material and the utilized extraction process. Employing a variety of treatment approaches results in significant variations of the shape, fibrillation degree, morphology, and properties of NFCs. A study performed by Desmaisons et al. has evolved an index of quality based on eight alternative criteria for measuring different reported NFCs

[48, 49].

## 2.2. Cellulose nanocrystals (CNC)

CNCs, also recognized as nanowhiskers, have a prolonged crystalline rod-like shape with superior rigidity to NFC due to greater removal of the amorphous domains. The typical crystallinity degree for a CNC varies from 54 to 88% [50, 51]. The source of CNC extraction is widespread, which includes garlic peel and cloves, *Posidonia oceanica*, wheat straw, coconut husk fibers, soy hulls, mulberry bark, banana rachis, pineapple leaf, grape skin, bagasse, rice straw, and sugarcane bagasse [52]. Various methods are employed for extracting relatively pure CNCs from the cell wall [23, 53]. Other methods such as high-intensity ultrasonication of microcrystalline celluloses (MCCs), microwave-assisted hydrolysis, acid hydrolysis, steam explosion, chemical pulping, and mechanical pulping (of wood) are also used to produce CNCs [54, 55]. Sulfuric acid hydrolysis or heat-controlled approaches present extraction processes, which are widely used for producing CNC [56, 57]. The intensive acid hydrolysis of cellulose fibers occurs through handling acid concentration, time, agitation, and temperature. This method results in the selective hydrolysis of non-crystalline cellulose domains and provides the CNCs with high crystallinity [27, 44]. Figure 2 shows conventional methods used for obtaining cellulose nanocrystal particles.

Along with acid hydrolysis, enzymatic hydrolysis and high-intensity sonication are alternative promising environmentally-friendly methods used to produce CNCs, which have gained much attention through the recent decades. As the high-intensity sonication takes away both the crystalline and amorphous regions of cellulose, this method is nonselective and results in the extraction of CNCs with lower crystallinity [58, 59]. It has also been declared that successive alkaline solution treatments at altered concentrations accompanied with mechanical treatments lead to the release of a significant quantity of nanocellulose with diameters ranging between 5 and 20 nm [60, 61]. Scientific reports also introduce other promising mechanical extraction processes, including waring blending, microfluidizer, grinding, and high-pressure homogenization [27, 62].

The properties of the extracted CNCs, such as the crystallinity degree, morphology, aspect ratio, and dimensional dispersity, are dependent on the conditions of reaction and the source of cellulose used [63]. It is known that the production of CNCs by sulfuric acid provides improved thermal stability, which is linked to the sulfate attachment on the surface of CNCs [64, 65]. Nevertheless, sulfuric acid utilization presents some disadvantages, including corrosion due to economic acid recovery, the presence of sulfate groups, and the dumping of a considerable quantity of salt obtained from the neutralization process of acid [66]. CNCs demonstrate various extraordinary properties, including superior strength, large surface area, high-magnetic response, and excellent crystallinity index [67].

The dimensions of CNC rods, also called whiskers, vary between 3 and 50 nm and 50 and 500 nm in width and length, respectively. CNCs present high elastic modulus (20–50 GPa) [68], high axial stiffness (105–168 GPa) [69], excellent tensile strength (~9 GPa) [70], superior thermal stability (~260 °C) [71], large aspect ratio (~10–70) [72], limited thermal expansion coefficient (~0.1 ppm/K) [73], low density (1.5–1.6 g/cm<sup>3</sup>) [74], lyotropic liquid crystalline behavior, and shear-thinning property [49, 75, 76].

Combining CNCs with other synthetic or natural polymers results in the emergence of functional composites. Bilodeau [77] and Kalia [78] have discussed various methods to produce CNC composites. Moreover, surface modification methods can change the self-assembly behavior of CNCs in suspensions and control their interfacial characteristics in composites. CNCs provide composites with improved mechanical behavior, decreased density, and increased surface area.

## 2.3. Bacterial cellulose (BC)

Bacterial cellulose nano-fibers (BCNFs) have a three-dimensional (3D) network of nanofibers and are derived from aerobic bacteria acting as an extracellular polysaccharide membrane by a bottom-up method [79, 80]. Bacterial cellulose (BC) presents a similar molecular formula to that of plants; however, it has a distinctive 3D micro- and nano-porous structure, which gives it excellent purity, increased polymerization degree, appropriate crystallinity (70–80%), great water content (90%), and proper thermal and mechanical stability [81, 82]. Nevertheless, BC costs significantly due to the utilization of expensive carbon sources and is not favorable in economic production [82, 83].

The lack of specific functional groups, aside from alcohol, and polymers, including pectin, lignin, and hemicellulose in BC, is the major distinction between BC and plant-derived CNFs [84]. Thus, BC is superior to other types of CNFs due to its good purity in the form of CNF, high crystallinity of about 80–90%, and great water-absorbing capacity [38, 85]. Bacterial nanocellulose (BNC) is made using cultivating bacteria in a water-based culture media, including nitrogen sources, carbon, phosphate, and glucose for a few days. Altering cultivation conditions, such as nutrient source, the ratio of oxygen, the type of bacterial strain, cultivation in a bioreactor, and incubation time, can modify the structure and behavior of BNC tubes [86–89]. Recent systematic studies have shown the influence of cultivation conditions on the BNC tubes and their properties [88]. *Gluconacetobacter xylinus* CGMCC No. 1186 was covered with fructose or glucose in bioreactors with a silicon tube. Studies have shown that fructose use improves the nanocellulose yield. Moreover, reactor variation results in altering the amount of dissolved oxygen and the structure of the achieved nanocellulose tubes [49].

Gatenholm and Klemm [90] have shown that a usual BC production process typically requires two weeks for cellulose production. Moreover, nanocellulose derived from BC presents superior crystallinity to nanocellulose produced from plants [91]. During several decades, the sources of cellulose production have largely changed. Until now, cellulose derivatives from some common bacterial types, including *Acetobacter xylinum* (*Gluconacetobacter xylinus* or *Komagataeibacter medellinensis*) and *Gluconacetobacter medellinensis*, have been studied due to their promising function as adsorbent, catalyst, and membrane [92]. Adsorbents, catalysts, and aerogel membranes based on BCs have been used for removing copper, lead, and dye, as well as the distillation of the membrane [93–95]. Overall, BC production aims to tackle the issues regarding its fermentation procedure, which demands novel low-cost culture mediums to enable BC production during decreased and limited times [38, 96].

Besides having the YOUNG's modulus of 78 GPa, reports have shown that BNC has an excellent capacity for holding water and a molecular weight of a maximum of 8000 Da [97]. BNCs are promising functional materials in application such as scaffolds and have excellent mechanical, optical, and magnetic properties—for example, the impregnation of in-situ Fe<sub>3</sub>O<sub>4</sub> nanoparticles for processing bacterial nanocellulose with magnetic properties or in the fabrication of ferromagnetic cobalt ferrite nanoparticles [98, 99] because of their inferior apparent density and large surface area [49, 100].

BNC is a suitable candidate for applications, such as implants and tissue engineering scaffolds, due to its simple processing for removing hemicellulose and lignin contaminants [49, 101, 102].

## 3. Synthesis of nanocellulose composites

The increasing necessity for biomaterials in industrial areas replaces traditional materials based on petrol. Nanocellulose has been combined with various materials ranging from either pure or composite ones [103].



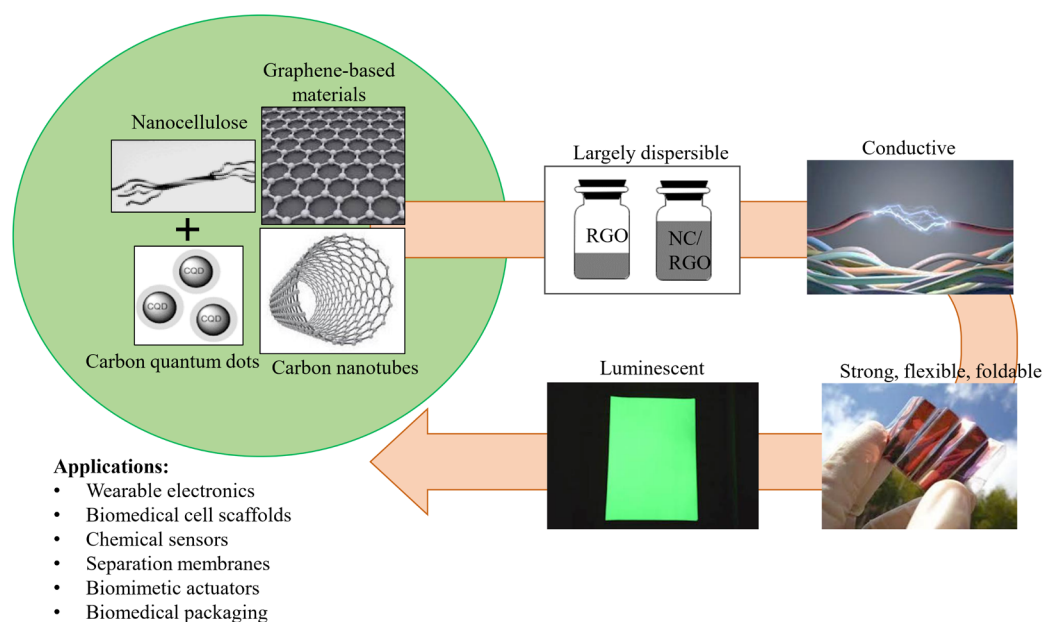


Fig. 3. The composite of nanocarbon and nanocellulose besides a few application areas and properties of these composites.

Nanocellulose provides a forum for implementing various materials on a nanometric scale owing to its large surface area, great porosity content, and excellent mechanical strength. Nanocomposites based on nanocellulose present mixed properties of the nanocellulose structure and the implemented nanomaterial with cooperative behavior [104]. Three main methods are generally used to implement nanomaterials into nanocellulose form, namely 1) adding and forming the implemented nanomaterial directly into a dispersed nanocellulose, 2) using BC membranes to form implemented nanomaterial inside the nanocellulose network, and 3) forming a layer of implemented nanomaterial directly on the surficial area of nanocellulose. Several implemented nanomaterials, including metallic nanoparticles (e.g. Pd, Ag, Au, Ag, Ni, CuO, and TiO<sub>2</sub>), as well as mineral-based nanomaterials (montmorillonite, SiO<sub>2</sub>, and CaCO<sub>3</sub>), and carbon-based nanomaterials (e.g. graphene and carbon nanotubes) are used to be implemented in nanocellulose materials. In terms of the utilization areas of the nanocomposites of nanocellulose, their use in environmental-related functions such as energy production, sensors, catalysts, and antibacterial materials are of great importance and consideration [49].

### 3.1. Nanocellulose–Nanocarbon Composites

Nanocellulose is merged with graphene-based materials, quantum dots of carbon, and nanotubes of carbon nanotubes, all of which are nanocarbons, giving practical characteristics to nanocellulose material (Fig. 3). Therefore, nanocellulose presents appealing characteristics such as biodegradability, biocompatibility, high mechanical strength, and non-toxicity, making them useful for applications in clothing due to being flexible, rigid, smoothly textured, reparability, and high deformation [105]. As a result, the general function of nanocellulose is to sustain nanocarbons, materials with demanding processing because of their high chance of forming aggregates. Composites of nanocellulose and nanocarbon present superior mechanical strength to those of polymer and nanocarbon [106]. Nanocellulose costs are higher than carbon-based nanotubes and graphene-derived materials. Using nanocarbons as dopants in the nanocellulose structure results in a limit of infiltration, which can provide electrical conductivity to the structure for lower prices than the pure nanocarbon. For producing composites of cellulose and nanocarbon, chemical approaches, such as surface modification, have been

used to enhance the solubility of cellulose [107]; however, it is also feasible to generate stability for dispersions without the employment of those methods. The advantageous interactions between the nanocellulose and nanocarbon reduce the chance of aggregation for the two materials [49, 108].

Applying the composites of nanocarbon and nanocellulose demands materials with excellent mechanical properties, biocompatibility, and electrical conduction. Separating nanocarbon into graphene-based materials, carbon nanotubes, and carbon quantum dots demonstrates the utilization area of composite materials [109, 110].

Nanotubes of carbon present elevated electrical and thermal conductivities, superb mechanical properties, excellent stability, and great aspect ratios. Adding these nanotubes improves the composites' mechanical strength and provides it with conductivity providing the presence of that an adequate quantity of connections. As cellulose and carbon nanotubes present great connections, they are known as natural companions [111]. The composite of cellulose and carbon nanotube has gained much interest in the areas of conductive papers [112–114] and fibers as well as wearable electronics [115], and aerogel [116], which has been only investigated in one study. Pure carbon nanotubes are likely to envelop cellulose fibers, resulting in the generation of a castable paper pulp presenting homogeneous electrical conductivity if made through a standard paper production process [115]. This composite paper illustrates the intervention of electromagnetics, which presents more efficient protection than circuit boards fabricated by printing metals. In addition, the addition of CNTs into cellulose leads to the enhancement of the stiffness and tensile strength of paper, which consequently creates a flexible and robust product. It is investigated that the ductility improvement owes to possessing long entangled and short components together: the nanocrystals of cellulose act as an inhabitant of the CNT aggregation and protect connections of the wide structure of nanotube [107]. Composites of carbon nanotube and cellulose are utilized as supercapacitor electrodes, biomimetic actuators, and biosensors by providing scaffolds with properties such as self-standing, lightweight, and foldability for titania [117] and polyaniline [49, 118].

Graphene presents special behaviors in terms of electronic carriage and electrocatalytic operations. Composites of cellulose and graphene are greatly porous and possess superb shape maintenance. These composites are used as sensitive and selective sensors of solvents, which

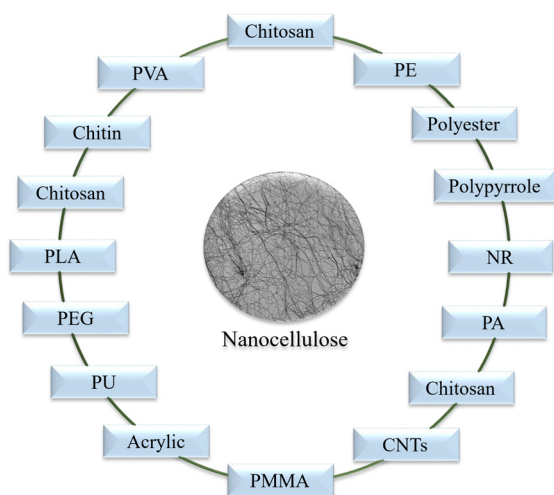


Fig. 4. A diagram of the latest studies on the composites of nanocellulose organic polymer.

operate based on observing capacitance change during adsorption [119]. According to the relative change of capacitance, the absorption of n-hexene, chloroform, toluene, ethanol, and acetone can be identified from each other [49].

### 3.2. Nanocellulose–Organic Polymer Matrixes

Nanocellulose in the form of fibers is largely used to strengthen polymer composites. Many studies have extensively investigated the function of the interphase layer on mechanical strength. Hydrogen bonding of parallel chains situated between the cellulose and crystalline areas provides the assemblage. Provided that a force efficiently conveys to the tough reinforcing domain, the elasticity modulus of the composite is the same as that of arbitrarily-oriented hard CNC because of the structure and the large surface area of the composites [49, 120].

Composites of nanocellulose and organic polymers have a sophisticated structure of high mechanical strength, intermolecular and intramolecular hydrogen bonds, and the capability of presenting a stiff, tight, and strong structure. Several polymeric matrixes, including epoxy resins [121], polyurethane [122], polystyrene-co-butadiene [123], polyethylene oxide-co-epichlorohydrin [124], polyvinyl acetate [125], and polystyrene-co-butyl acrylate [126], have been strengthened by nanocellulose. These organic composites are fabricated with the aid of a range of approaches, such as impregnation of the solution [127], hot pressing, freeze-drying, [128], and compression molding [129], among which casting is widely employed [130–132]. The latest studies on the composites of nanocellulose organic polymer, besides their application areas, are illustrated in Figure 4.

Two varieties of approaches are generally used for producing cellulose-based composites: (1) the two-stage approach, which involves cellulosic solution achieved through the full dissolution of a cellulose portion that is strengthened by substitutional cellulose, and (2) the one-stage approach through which an incomplete dissolution of cellulose generates a matrix domain sedentary nearby the residual fiber center [49, 133].

The optical, mechanical, and thermal properties can be improved with the implementation of nanocellulose in the polymeric matrix. For instance, Tan et al. [134] recently fabricated a uniform cellulose fiber dispersion in coatings of waterborne acrylic that presented enhanced transparency and characteristics. The coatings made of composite demonstrated the utmost increase of 500% in elastic modulus, doubled hardness, and a decrease in abrasion loss by 35% compared to the properties of the pure coating. To produce high-quality materials, nanocellulose reinforcements have been used with thermoplastic (polyethylene oxide (PEO), polylactic acid (PLA), polyvinyl alcohol (PPA), polypro-

pylene carbonate (PPC), polypropylene (PP), and polyethylene (PE)) and thermoset (polyurethane and epoxy) polymers.

### 3.3. Nanocellulose–Inorganic Nanoparticle Composites

Owing to the cooperative result of the mixed chemical and physical interactions between the inorganic and organic elements, hybrid organic-inorganic nanomaterials usually present enhanced physicochemical properties such as optical, mechanical, and thermal properties, together with conductivity [120, 134]. Novel materials have been introduced due to incorporating organic strengthening elements such as nanowhiskers and nanofibers of cellulose in a range of composite matrixes [49].

Hybrid composites of nanocellulose and inorganic materials are widely prepared by atomic layer deposition, dispersion of doping casting ionic liquid/cellulose, layer-by-layer deposition, and liquid phase deposition. Hybrid composites of metal nanoparticle and nanocellulose are typically synthesized through reduction by either an extraneous agent, a reduction functional group implemented on the superficial area of nanocellulose, or the inherent reduction of the hydroxyl and aldehyde groups of nanocellulose [109]. In a remarkable study, mixing CNCs and Ag wire exposed to light resulted in the formation of small, catalytic Ag nanoparticles on the surface of CNCs. The light plays a crucial role in dissolving the Ag wire and provides a specific concentration of  $\text{Ag}^+$  ions in the solution. Carbon–oxygen double bonds are the products of the hydroxyl group oxidation on the surface of CNC, while the silver is reduced on the exterior areas of CNC. Nanoparticles of ruthenium were fabricated through a mixture of the chemical oxidation and CNC surface, which are extremely challenging to fabricate from usual ruthenium(III) chloride ( $\text{RuCl}_3$ ) [135].

## 4. Application of nanocellulose composites in the environmental engineering

Nanocellulose is a promising material in packing, electronics, optoelectronics, pharmaceutical materials, biomedical applications, nourishment, and cosmetic materials due to its several extraordinary benefits, including excellent water absorption, non-toxicity, maintenance, biocompatibility, and high mechanical strength. More importantly, nanocellulose separated from a range of biomass junks presents cost-effectivity relative to other raw materials. As a result, the advent of valuable produce through nanocellulose derived from biomass junk materials enhances the economic status and protects nature [136–138].

The nanocellulose-based materials prepared through the previously stated approaches possess characteristics such as recyclability, sustainability, and carbon neutrality [102]. As a result, their characteristics turn these materials into environmentally-friendly nanomaterial, which present a favorable function when employed in composites. Combining these materials with inorganic and organic substances results in the production of a composite, which can be extensively employed in optoelectronic, electronic, detection of human motion, packaging, and many other applications [103].

### 4.1. Application of nanocellulose composite in the catalysts

The use of nanocellulose composites is increasing for treating polluted water through the catalytic activity of organic contaminants [139]. Two types of implemented nanoparticles are applied for catalytic purposes, which are photocatalysts such as titanium dioxide ( $\text{TiO}_2$ ) and precious metals including gold (Au), silver (Ag), and platinum (Pt). Generally, nanocellulose catalysts sustain the catalyst for prohibiting the aggregation of nanoparticles [15].

Combining microfiber carbons (MFCs) and  $\text{TiO}_2$  nanoparticles would result in the formation of transparent and stiff thin layers. FTIR

results have presented that  $\text{TiO}_2$ -NPs and the surface of MFC interact electrostatically with each other. Increasing the amount of  $\text{TiO}_2$  lowers both the mechanical strength and the transparency of the nanocomposite [140]. It is suggested that this nanocomposite may be applied for photocatalytic applications to decompose organically-based pollutants; however, this suggestion has not been clearly investigated here. There are some huge proofs regarding this suggestion in articles. To name a few, a study claimed that a nanocellulose-based aerogel was coated with a  $\text{TiO}_2$  layer, with a thickness of 7 nm, through a chemical vapor deposition method. The pollutant was modeled with methylene blue, which was effectively decomposed afterward under UV light, suggesting that the material is promising for treating water [141]. Besides, the nanocomposite of  $\text{TiO}_2$  and nanocellulose is an excellent photoswitch superabsorbent of water. The cellulose aerogel coated with  $\text{TiO}_2$  presents hydrophobic behavior (contact angle of  $140^\circ$  for water). The UV irradiated nanocomposite shows super hydrophilic characteristics, and by keeping it in the dark, the composite becomes hydrophobic again [141]. The MFC aerogel coated with  $\text{TiO}_2$  has the potential to be utilized for absorbing spilled oil from water as it has extremely lightweight and hydrophobic behavior, enabling it to suspend on the surface of the water and take in organic solvents up to almost 20–40 times of its weight [20].

Precious metals, including Pd, Pt, Ag, and Au, are widely used for catalytic activities. Nanocomposites of metal nanoparticles and nanocellulose are mainly fabricated through metal salts reduction when presented to nanocellulose. Various chemical materials, including hydroxyl groups of nanocellulose,  $\text{Na}_3\text{citrate}$ , sodium borohydride ( $\text{NaBH}_4$ ), and hydrogen ( $\text{H}_2$ ), are employed as reducing agents. In a study, Cirtiu et al. put a suspension of palladium chloride ( $\text{PdCl}_2$ ) and NCC into an atmosphere of  $\text{H}_2$  with a pressure of 4 bar for 2 h. This caused the  $\text{Pd}^{2+}$  to reduce to palladium nanoparticles (PdNPs) with a particle size of about  $3.6 \pm 0.8$  nm. Cyclohexanone was formed as a product of phenol transformation through the hydrogenation activity of catalysts on the nanocomposite of PdNP and NCC. The mentioned reaction did not happen by synthesizing Pd nanoparticles without the presence of NCC. The outcomes and products are ascribed to PdNPs aggregation followed by a decrease in responsive surface area when there is a lack of the covering role of NCC [142].

Likewise, the production of dispersed AuNPs was supported using MFCs, which were oxidized by TEMPO. Through the synthesis process, the reduction of MFC surface covered with  $\text{Au}^{3+}$  occurred using  $\text{NaBH}_4$  at ambient temperature, resulting in the formation of extremely dispersed AuNPs. The dispersion quality of AuNPs crystals was confirmed with the aid of TEM and selected area electron diffraction (SAED) data, which displayed convenient dispersion of crystals with a homogeneous diameter of 5 nm situated mainly along the fibers. The nanocomposite of AuNP and MFC demonstrates a UV-vis absorption spectrum, which is not as broad as those of the composite of AuNP and cellulose or the solution of AuNP, suggesting that the nanocomposite of AuNP and MFC includes lower aggregated Au nanoparticles. The author hypothesizes that the carboxylate groups of MFC stabilize Au nanoparticles and hinder the aggregation of Au nanoparticles. The nanocomposite of AuNP and MFC demonstrated high catalytic activity in 4-nitrophenol degradation. Nitroaromatic-based materials, undesired organic derivatives of pharmaceuticals, agrochemicals, and dyes, present toxicity [15, 143]. Thus, 4-nitrophenol is utilized mainly as an example for investigating the catalytic function of metal nanoparticles [143–145]. The nanocomposites of AuNP and MFC presented site time yields of more than 840 times higher than those of the traditional composites of AuNP and polymer [145]. An environmentally-friendly approach was introduced by Wu et al. to synthesize the nanocomposites of AuNP and NCC. The synthesis method was carried out at an ambient temperature, during which the sedentary reduction of  $\text{Au}^{3+}$  using NCC hydroxyl groups resulted in the formation of 30 nm-diameter AuNPs. Data from TEM confirmed the

well-distribution of the AuNPs and indicated that the nanoparticles were not grown along the fibers of NCC. The resulted nanocomposites of AuNP and NCC were far more effective, up to 3 times than the AuNPs, without supporting through the degradation of 4-nitrophenol by catalysts because of the improved AuNPs dispersion [144].

Azetsu et al. [146] also studied the catalytic performance of Pd and Au bimetallic nanoparticles through cellulose MFCs, which were oxidized by TEMPO acting as a supporter. Because of the great carboxylate groups' density, which existed in the MFCs with concentrations of 1.0 wt% and 0.96 mmol/g (carboxylic acid ( $-\text{COOH}$ )), nanoparticles of hybridized Au–Pd, Pd, and Au with catalytic behavior and excellent dispersion were sedentarily produced on the surface of MFCs. The reduction agents of  $\text{NaBH}_4$  and the anchors of AuNP of carboxylate groups were employed in this method. A variety of NP and MFC nanocomposites have been compared regarding their catalytic activity for reducing 4-nitrophenol to 4-aminophenol. The site time yields of the reaction, including reduction and catalysts, were assessed through the absorbance change at 400 nm. The results demonstrated that the Au nanoparticles with a size of 4 nm, which were deposited on the MFCs, illustrate greater catalytic behavior than a condition in which the nanoparticles are deposited on other types of natural or synthetic polymers.

Nevertheless, greater catalytic activity in 4-nitrophenol reduction is assigned to the hybrid nanocomposite of Au–Pd NP and MFC (ratio of Au: Pd equal to 3:1). Some suggestions state that the improved catalytic performance may arise from the impacts of electronic mediated ligands. These effects of electronic-mediated ligands speed up the reactions of catalysts prompted by electronic interactions occurring between two metal nanoparticles concerning the single nanoparticles of metal.

Copper(II) oxide ( $\text{CuO}$ ) nanoparticles are additionally employed as catalysts besides  $\text{TiO}_2$  and precious metal nanoparticles. Through the 4-nitrophenol degradation by  $\text{CuO}$ -NP/NCC catalysts, the reduction of  $\text{Cu}^{2+}$  via  $\text{NaBH}_4$  at room temperature occurred and resulted in the formation of  $\text{CuO}$  nanoparticles on the surficial area of NCC. The nanocomposites of  $\text{CuO}$  nanoparticles and NCC nanocomposites presented superior function for the degradation of 4-nitrophenol to  $\text{CuO}$  nanoparticles, which were supported by graphene or were not supported at all. This is due to the greater surface area of NCC and the immobility of  $\text{CuO}$  nanoparticles using NCC hydroxyl groups [143]. All the studied literature in this part declare that the deposition of metal nanoparticles on the porous NCC is random and the covering percentage of the surface of nanoparticles on NCC is limited. Therefore, Padalkar et al. suggested applying cetyltrimethylammonium bromide (CTAB) to enhance the covering percentage of the surface metal nanoparticles on NCC. The adsorption of CTAB on the surface of metal nanoparticles, through its cationic quaternary ammonium groups, could present interaction with hydroxyl groups that are high in electron levels as well as anionic groups on the NCC surface. In contrast with the synthesis condition in the absence of CTAB in which the metal nanoparticles formed on the surface of the TEM grid, the synthesis procedure with the use of CTAB resulted in the formation of metal nanoparticles along the fibrils [15].

#### 4.2. Application of nanocellulose composite in the pollutant sensors

Composites of nanocellulose thin layers and phenolic resin have superior average shear strength ( $\sim 9.6$  MPa) to that of non-nanocellulose Cu–Cu joints ( $\sim 4.7$  MPa) [147, 148]. The thin composite layers based on bacterial nanocellulose presented considerably high elasticity modulus (28 GPa) compared with fibrillated kraft pulp composites because of a greater aspect ratio, purity, crystallinity, and homogeneous size [149].

Leal et al. [150] fabricated a composite film made up of cellulose nanocrystals via tapioca starch and glycerin as precursors. The prepared film of the composite showed an appropriate barrier of oxidation and mechanical performance. The penetrability of water vapor was 2.5-folds

lower, and the maximum stress was about twice the starch film with the absence of nanocellulose. The prepared film of composite demonstrated its promising performance in the food packaging industry.

In a study, a supercapacitor was made up of nanocellulose and CNTs with possessing the capacitance of 14.9–16.5 mF and series resistance of 74–155  $\Omega$  at a 1.8 cm<sup>2</sup> section. This illustrates far superior capacity for storing energy to the films of CNT films (10mF and 30 $\Omega$ ) with a great power density and extended life duration. The mentioned inexpensive products used as storage can be extensively applied in self-governing intelligence technology [151]. Nanocellulose composites are alternatively used in vehicle, aerospace, electrochemical cells, and sewage treatment industries [152].

Many scientists have evolved biosensors based on the nanocomposites of Au nanoparticles and nanocellulose [153]. In order to recognize chemicals with electrochemical routes, some researchers used the AuNP conductivity and the biocompatibility of BC through incorporating the nanocomposite into an electrode [154–156]. Zhang et al. [155] prepared nanocomposites of Au nanoparticles and BC for hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) sensors and glucose. At the first stage, ultrasonication was employed to disperse BC hydrogels in water, followed by the addition of polyethyleneimine (PEI) and HAuCl<sub>4</sub>. During the next step, the compound was held at 60 °C when the formation of Au nanoparticles occurred on the nanocellulose surface with PEI acting as a linking and reducing agent. The addition of a variety of halides to the mixture changes the golden shell on the surface of nanocellulose. SEM images presented that adding chloride triggered a homogenous and even coating of Au nanoparticles, with a size of 9 nm, on the surface of the BC surface. The addition of bromide provides agglomerates of Au nanoparticles, which are greater in size and superior covering of Au nanoparticles on the surface of BC. Adding iodide provides submicron Au nanoparticles and an inferior covering of Au nanoparticles on the surface of BC. The nanocomposite of Au nanoparticles and BC obtained from this method was coated on a glassy carbon electrode (GCE) followed by being functionalized with myoglobin (MB), hemoglobin (HB), and horseradish peroxidase (HRP). The as-prepared AuNP/BC/MB/GCE, AuNP/BC/HB/GCE, and AuNP/BC/HRP/GCE were shown to present effective performance as sensors for H<sub>2</sub>O<sub>2</sub> while AuNP/BC/HRP/GCE demonstrated the most effective function [154, 155]. At the same time, the composite of AuNP/BC/GCE functionalized with the glucose oxidase (GOx), and HRP highlighted the best application as a sensor for glucose [156].

Au and Ag nanoparticles have been widely investigated by surface-enhanced Raman spectroscopy (SERS) [157–160]. The biosensors prepared based on this approach are employed in studying organic pollutants and aquatic pathogens. For instance, former research performed by our group observed two known aquatic protozoa, *Cryptosporidium parvum*, and *Giardia lamblia*, by a biosensor based on an immunogold. As directly applying the colloids of AuNP and AgNP is usually unsuitable in real-life circumstances, a flexible substrate based on SERS-like paper is suitable. Nanocellulose could be used to prepare films and papers to host nanoparticles activated by SERS. Despite that, the function of the nanocomposites of Au nanoparticles and nanocellulose or Ag nanoparticles and nanocellulose for SERS underlayers have been investigated only in a handful of researches [161, 162]. Marques et al. [162] prepared the nanocomposite of Ag nanoparticles and BC by in situ Ag<sup>+</sup> reductions using Na<sub>3</sub>Citrate on the matrix of BC.

The produced substrate of Ag nanoparticle and BC presents the ability to recognize 2,2'-dithiodipyridine and thiosalicylic acid at concentrations of 10–4 M. The SERS spectra of the L-histidine, L-glutamin, and L-phenylalanine amino acids have been achieved using this substrate. Park et al. [161] also synthesized the nanocomposite of Au nanoparticle and BC to act as an underlayer for the detection of phenylacetic acid (PAA) and 4-fluorobenzenethiol (4-FBT) trace organic chemicals based on SERS. This method benefited from the 3D structure of BC.

Deformation of the 3D nanocomposite of Au nanoparticle and BC rises from the drying process generates hot spots along the vertical direction, which causes far more Raman signals than conventional 2D SERS substrates. Au nanoparticles could alternatively be functionalized to obtain other aims than their direct application in the biosensors. Pinto et al. performed the deposition process of the nanoparticles of Au on BC via Au<sup>3+</sup>–BC complex reduction using Na<sub>3</sub>Citrate. A uniform SiO<sub>2</sub> coating on the nanoparticles of Au was evolved for specifically separating the nanoparticles of Au. Using this method, the thickness of the SiO<sub>2</sub> shell on the nanoparticles of Au could be altered in the range of 15–100 nm, which has also been noticed in TEM results. The membranes of BC, which were functionalized using the nanoparticles of Au or Au–SiO<sub>2</sub>, were fabricated afterward through layer-by-layer (LbL) deposition with the aid of PSS, poly(sodium 4-styrenesulfonate), PDDA, and poly(diallyldimethylammonium chloride). This study proposes that it is likely to curb the optical behavior of the nanocomposites of Au nanoparticles and BC via covering the Au nanoparticles with SiO<sub>2</sub> and by varying the constitution of the layers with the membranes of Au–SiO<sub>2</sub> nanoparticles and BC alongside Au nanoparticles and BC. One of the convenient utilization areas of this nanocomposite is a core-shell biosensor based on paper if long-run chemical and stability are of great importance [163]. Nanocomposites of Au nanoparticles and NCC can be used as a substrate for immobilizing enzymes. At the first step, Au nanoparticles are generated on the surface of NCC by reducing the HAuCl<sub>4</sub> using NaBH<sub>4</sub>. Afterward, thioctic acid (Thc), including –SH and –COOH, binds to the surface of Au nanoparticles and its carboxylic acid groups activated through N-hydroxysulfosuccinimide (NHS) and 1-ethyl-3-[3-(dimethylamino)propyl]carbodiimide (EDC). Eventually, the amino groups of the enzyme – cyclodextrin glycosyltransferase (CGTase) – make covalent bounds with the activated carboxylic acid groups on the surface of nanocomposites. The capacity of CGTase on the nanocomposites of Au nanoparticles and NCC for making bounds is 165 mg<sup>−1</sup> of NCC, and the CGTase presented high biocatalytic performance [164]. The significant amount of enzymes on the surface of this nanocomposite implies that it is useful for producing biosensors based on enzymes [15].

#### 4.3. Application of the nanocellulose composite in the flocculant

Flocculants based on biopolymers including alginate, cellulose, tannins, and chitosan, have gained much attention. These materials are famous for their biodegradability, having a large specific surface area, and possessing nanometric dimensions [165, 166]. Generally, there are two types of natural polymer flocculants [167]. The first type includes natural polymers grafting to produce semi-natural flocculants such as polyacrylamide-grafted hydroxypropyl methylcellulose [168]. The second type is obtained via direct modification of natural polymers to produce enhanced natural flocculants, including dicarboxylic acid nanocellulose [169]. Studies regarding the utilization of nanocellulose chemicals for water are not enough as scientists' major consideration is on metal adsorption from thinned watery solution or photocatalysis as mentioned previously. One of the challenges regarding the utilization of CNCs as flocculants is agglomeration, which is because of the presence of hydrogen bond networks originating from the –OH groups on the surface of CNCs [170].

Nevertheless, the characteristics of CNC flocculants may alter by changing the –OH groups on the surface, thus, the appropriate quantity of –OH groups on the surface should be recognized for preparing a useful flocculant. Anionic surface groups, including hydroxy groups, are widely observed in nanocelluloses following the pulping process because of the plant cell wall components [171]. As a result, anionic nanocellulose and a range of inorganic minerals are typically negatively charged and weak in interactions. Furthermore, reports have stated that an anionic flocculation agent has inferior solubility in acid-based



solutions, hindering its functional use as a flocculant in the mentioned states [14].

A range of chemical pretreatments, including citric/hydrochloric acid hydrolysis [170], 2,2,6,6-tetramethylpiperidin-1-oxyl-mediated (or TEMPO-mediated) [172], aminoguanidine-reacted wood celluloses [173], periodate-chlorite oxidation [171, 173], and carboxymethylation [171], have been employed to increase the density of anionic charge and bestow nanocellulose with cationic charges to extend its application. Nanocellulose adjustment for preparing cationic equivalents is significantly beneficial for removing anionic particles via molecular interactions occurring between different charges that lead to the aggregation of particles in applications such as wastewater treatment [14].

At this time, it is possible to functionalize nanocellulose for creating efficient flocculants through the subsection of hydrophobic, cationic, or anionic functional groups to the surface of nanocellulose by the neutralization of charge. The surface of cellulose, activated by  $-OH$  groups, stimulates effective nanocellulose functionalization, adds proper functionality, and provides largely efficient flocculants [174]. Furthermore, presenting functional groups to the cellulose can improve its interaction with various compounds to boost the polarity and hydrophilicity of the surface [14].

Suopajarvi et al. studied a range of functionalized nanocelluloses, including cationic and anionic dialdehyde, which have been used for flocculating wastewater. Their study outcome was hopeful, demonstrating that functionalized nanocellulose could be used as an agent for treating wastewater. Presenting anionic groups (e.g. carboxy) detaches the network of the nanocellulose surface groups through hydrogen bonding, which subsequently enhances the nanocellulose performance as an adsorbent [139]. For example, interval oxidation is used for implementing aldehyde groups by oxidizing surface groups of  $-OH$  and varying the carbonic structure of the glucopyranose ring [175]. The mentioned 2,3-dialdehyde cellulose aldehyde groups are facile to be subsequently transformed into different functional groups, such as imines, sulphonates, or carboxylic acids [171]. The application of anionic and cationic dialdehyde celluloses, obtained by aqueous interval oxidation, has been researched by Liimatainen et al. [176, 177] who demonstrated that the nanoparticles of anionic cellulose provided an improved flocculation function of kaolin compared with materials based on cationic dialdehyde cellulose.

Oxidizing nanocellulose using TEMPO in the presence of water with moderate conditions is an additional useful approach for adjusting nanocellulose in chemical ways for generating a significant number of charges. The mentioned approach effectively converts surface hydroxymethyl groups to their related types of carboxylic [172]. Then, a nanocellulose aggregation can be hindered by negative charge groups presented on the surface of nanocellulose through repulsive forces. Chen et al. [178] presented NCC materials stabilized by electrostatic forces that could be prepared through a reaction including three steps: the oxidation of periodate, chlorite, and TEMPO. The researchers stated that periodate oxidation degraded cellulose, which could be limited through minimizing the oxidation of periodate and increasing the oxidation of TEMPO [14].

Many researchers have investigated the chemical and physical modification of the nanocellulose structure to enhance its behavior and characteristics. To name a few, Sun et al. [179] studied the influence of CNC application on the flocculation of Gram-negative bacteria (*Pseudomonas aeruginosa* PAO1). This research presented that the output of the bacteria-depleted flocculation relied on the form of the particles of cellulose colloids. Furthermore, the research demonstrated that particles having the form of a rod, which is normally seen in CNC, could greatly deplete colloids compared with particles with the form of a sphere. Moreover, the competence of the flocculation – flocculation of anionic microalgal cells - of CNCs increased by presenting imidazole and cationic pyridini-

um functional groups via the grafting method in comparison with that of typical polymer-based flocculants [174, 180]. All in all, many scientists have studied a range of methods for modifying nanocellulose to expand its usage in the industry [14].

#### 4.4. Application of nanocellulose composite in energy fields

Lately, several studies have prepared appliances for storing energy with BNC [181]. Besides these apparatuses, numerous developments have also happened in the environment and technology [182–185]. BNC can be applied for strengthening materials or is being reinforced by additives based on its utilization. Recent studies have tried to fabricate advanced flexible BNC-based materials with no impact on their intrinsic characteristics [186]. Many researchers have investigated cellulose-based materials as a promising alternative. Many investigations have also been performed regarding applying a combination of BNC and CNTs [187]. The studies experimented with the electrochemical efficiency and function of BNC nanocomposite electrodes with their unique 3D porous structure, which makes them a promising material for storing usages [185]. The NFC is a valuable material for several applications, including printed electronics, electronic gadgets, sensors, and smart electronic materials. Its various properties and characteristics include mechanical strength, limited thermal expansion, transparency, inferior surface roughness, and ignorable surface roughness. In addition, NFC has been widely used in nanocomposite fabrication with amylopectin, polyurethane, styrene butyl acrylate, melamine-formaldehyde, and phenolic resin. [188–190]. The underlayers based on NFC are efficient in fabricating electronic gadgets for substituting conventional high-cost glass preparing methods. NFC has more enhanced heat transfer behavior than glass. Moreover, the transparency of NFC arises from its closely packed structure and a minor fiber space inhibits the scatter of light [191].

Nanocomposites of nanocellulose could be used in fabricating Li-ion batteries, solar cells, and fuel cells. Fuel cell function is to produce electricity from chemical energy. Fuel, normally  $H_2$ , oxidizes via catalytic activities at the anode while  $O_2$  is reduced catalytically to  $H_2O$  at the cathode. Through an employed circuit, the electrons are transferred from the anode to the cathode. BC is a suitable underlayer to gather a considerable quantity of anode catalysts with nanometric size. By immersing the BC membrane in a solution with the ammonium hexachloropalladate concentration of 5 mM, precipitation of Pd nanoparticles forms in the BC membrane.

Nevertheless, immersing plant cellulose in hexachloropalladate solution does not lead to the formation of precipitates. With the aid of tapping mode AFM, it was found that the structure of plant cellulose was weaved closely together with organized fibers; however, the fibers of BC fibers had a tunnel-shaped structure, which was not as dense as the former one. Consequently, it is claimed that reducing groups inside their loose structure are responsible for the precipitation of Pd nanoparticles in BC. In a study, a native membrane of BC was subjected between the layers of the nanocomposites of PdNP and BC to be applied in a fuel cell as a membrane electrode assembly (MEA). The density of Pd in the cathode and anode membranes was  $0.4 \text{ mg/cm}^2$  in dry weight conditions, while the MEA thickness was  $150 \text{ }\mu\text{m}$ . The oxidized  $H_2$  was employed to the PdNP/BC anode, and the greatest current was around  $0.26 \text{ mA}$ , which presents that this nanocomposite is a promising one in applications for converting energy. The mentioned membrane of PdNP/BC demonstrates more benefits than alternative polyelectrolyte-based membranes because of its limited gas crossing and enhanced thermal stability [192].

Solar cells are used to generate electricity from solar energy. Due to the high mechanical strength, smoothness, and transparency of nanocellulose papers, they can be applied efficiently for producing the substrates of solar cells. Hu et al. [193] devised a solar cell with transparent papers

of nanocellulose using an indium-tin-oxide (ITO)-coated paper of nanocellulose, which was prepared via radio frequency magnetron sputtering with a target made up of 90% of  $\text{In}_2\text{O}_3$  and 10% of  $\text{SnO}_2$ . Followed by the coating process, the transparent paper turned translucent. Afterward, the coated paper of nanocellulose underwent treatment by [6:6]-phenyl-C61-butyric acid methyl ester (PCBM), with poly(3-hexylthiophene) (P3HT) acting as a layer for absorbing light. The application of this transparent paper lengthened the light path inside the absorbent layer, which led to more absorption of sunlight. The fabricated solar cell presented an efficiency of 0.4% for power conversion, demonstrating the potential of nanocellulose papers to be applied in photovoltaic devices. Nonetheless, the short-circuit current and total power conversion efficiency (PCE) of the solar cell made from nanocellulose papers are inferior to those of the glassy solar cell due to the ITO sheet resistance on nanopaper, which is about 50–100 times more than that on glass.

Besides solar cells and fuel cells, nanocellulose can alternatively be employed in Li-ion batteries (LIBs), which are rechargeable energy storage appliances, very reputable, and are extensively applied in customer electronic devices. Nanocellulose papers make it possible to fabricate functional and flexible LIBs with a limited thickness. Jabbour et al. have thoroughly reviewed cellulose LIBs [194]. In this study, we discuss LIBs based on nanocellulose and their benefits. Some reports have shown the application of nanocelluloses for electrolyte, separator, and electrode in LIB cells [195–198]. Jabbour et al. prepared a flexible LIB anode based on graphite/MFC by performing vacuum drying on a slurry of graphite/MFC at 313 K for several hours. Besides the anode of graphite/MFC, the cell involved the electrolyte of diethyl carbonate and ethylene carbonate together with LiPF<sub>6</sub> and a cathode of Li foil. The anode based on graphite/MFC presented the same charge/discharge capacity as the typical graphite/poly(vinylidene fluoride) (PVDF) plus suitable stability, illustrating that tenable MFCs could substitute the synthesized chemical PVDF to fabricating LIB anodes [199].

Hu et al. [200] also fabricated a nanocellulose paper combined with CNT, coated with Si (Si-nano paper) to be applied as a LIB electrode. Initially, the paper based on CNT/nanocellulose was fabricated through freeze-drying of the uniform mixture of a hydrogel from CNT/nanocellulose. The CNT implementation in the composites provides improved electrical conductivity. In the next step, the plasma-enhanced CVD (PECVD) approach was utilized to deposit a low-thickness Si-based coating on the paper of CNT/nanocellulose. In the final step, the cell was fabricated using Li metal foil, Si-nanopaper, and a Celgard 2250 separator immersed in an electrolyte involving LiPF<sub>6</sub> in ethylene carbonate/diethyl carbonate. The fabricated Si-based nano paper presented a persistent capacity equal to 1200 mAh/g after 100 cycles of charge and discharge, which is more than three times higher than the theoretical capacity for a typical graphite-based anode. In addition, the flexible and significantly porous structure of the paper of CNT and nanocellulose, introducing plenty of housing locations for the expansion of the Si layer during lithiation, provides the Si-based nanopaper with enhanced stability.

Leijonmarck et al. [198] incorporated the MFC separator and electrode into a paper to fabricate a LIB cell with suitable flexibility. The desired cell was produced using successive vacuum filtering of the negative electrode suspension, which involved MFCs and graphite together with a separator suspension made up of MFCs and  $\text{SiO}_2$  as well as the positive electrode suspension, containing MFCs and  $\text{LiFePO}_4$  on a filtering paper. The dehydrated cell includes three layers mounted on each other as well as two outer electrodes and one centrally-positioned separator. This flexible paper-based battery possesses high strength and demonstrates appropriate capacity cycles of charge and discharge. The MFCs play a crucial role in bonding the parts and supplying a network with enough strength and flexibility for the LIB cell. Apart from nanocellulose application in separators or electrodes, they could be employed

in electrolytes to strengthen and reinforce the device.

## 5. Conclusions and future insights

This study concisely investigated the late developments in the fabrication of nanomaterials based on nanocellulose and their prospective utilization in environment-related areas. The sources of nanocellulose are vast and inexpensive. The top-down cellulose pulp decomposition leads to the fabrication of highly thin nanofibers possessing diameters of 2–5 nm. These nanocelluloses present unique thermal, mechanical, and optical behaviors. Aerogel, fiber, film, and papers of nanocellulose fabricated through the bottom-up method present enough location to act as a powerful bearer for integrating implemented nanomaterials. The nanocomposites based on nanocellulose present several benefits, such as catalytic and antimicrobial activities, which enable the materials to be used in water treatment. Nanocellulose combined with Au nanoparticles could be a suitable SERS underlayer, provided that the size and loading of Au nanoparticles are improved. In addition, nanocellulose is utilized as a modern substrate for the fabrication of fuel cells, presenting promising usage in energy devices.

Over the following years, approaches for ameliorating the distribution and size of metal and other nanoparticles in nanocellulose underlayers regarding their utilization will gain much attention. Fine and well-dispersed nanoparticles are beneficial for catalyst areas, whereas large and aggregated metal nanoparticles present a better performance in the SERS area. The loading of nanoparticles is additionally pretty paramount in nanocomposites. Investigating the loading limitation and quantity is crucial for various nanoparticles in the matrix of nanocellulose and requires further study. Furthermore, incorporating inorganic material additives in the nanocellulose can change their durability. Consequently, studies have to try to extend the life of nanocomposites or refabricate them in order to turn them into durable materials. Nanocellulose nanocomposites are promising materials as a large number of researchers have searched for finding a low-cost and green material to fight against today's environment-related issues.

Until now, the majority of review studies have concentrated on only a distinctive kind of nanocellulose: aerogel sorbents, adsorbents, or a special kind of treatment for ecological recovery such as water treatment. The present review focuses on introducing an outline of the latest advances and future trends of a range of nanocelluloses applied in various fields of environmental recovery and treatment, such as previous outcomes, contemporary development, and future trends. The latest advances of nanocellulose employed as a membrane, flocculant, photocatalyst, or adsorbent for different utilizations in environment recovery and treatment are discussed thoroughly here.

This review emphasized studies and developments in the fields of NCC, NFC, and BNC. Future probable usage of these materials is varied, ranging from medication and biomedical areas to marketable commercial productions. Choosing the best nanocellulose for a specific application is demanding and vital. In the pharmaceutical and drug delivery fields, BNC is the best candidate. Nevertheless, the NFC is a perfect choice in areas such as rheology qualifiers and additives of composites, which require a larger amount of materials. On the other hand, NCC is ideal to be used in applications including the biomedical field, rheology qualifier of polymer composites, and interface stabilizers. Several studies performed by different researchers and companies have explored the production enhancement and commercial development of nanocellulose.

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## Conflict of interest

The authors declare that there is no conflict of interest.

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