Chapter 5
Functionalization of Cellulose-Based Materials

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Abstract Multifunctional hybrid materials based on nanocellulose have gradually emerged as a substitute for petroleum-based materials. In this chapter, we briefly present the latest technology in this field, including processing, functional properties, and areas of application. For example, the combination of cellulose nanocrystals (CNCs) with different types of organic or inorganic nanoparticles enables the study and analysis of multifunctional nanohybrids with important scientific and industrial applications and opens new horizons in materials science. In particular, technical analysis, including supercapacitors, solar cells and batteries, separation technology, and wastewater treatment, catalysis.

5.1 Introduction

Cellulose comes from a variety of sources, can be derived from plants and is a renewable, bio-based polymer. A new class of renewable crystal particles, represented by nanocellulose, can be combined with other micro- or nanomaterials, enabling the development of efficient and sustainable multifunction devices (Klemm et al. 2018). Research into CNC may require improvements in a variety of projects, from platforms or supercapacitors for biomedical applications, permeable separators for batteries, slim films for hardware and anti-counterfeiting applications, for detection, for standalone catalytic membranes or water disinfection applications (Lizundia et al. 2017a, b; Rescignano et al. 2014).

To move from the laboratory to commercial and modern applications, specialists should complete the transition from removing CNC from research facilities to modern scale creation (Reid et al. 2017). Currently, there are many organizations that are

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able to transport kilograms to large quantities of nanocellulose per day using various flushing systems. Nevertheless, unique extraction strategies can greatly influence CNC strength and surface science, surface charge thickness, and molecular size (Foster et al. 2018). Therefore, it is essential to use key and standard testing techniques to characterize these properties to ensure reliable material execution. In general, the current level of modern production of cellulose nanocrystals is remarkable, which offers incredible potential for commercial applications.

As noted by Stratistics Market Research Consulting, the global CNC market size will reach $22.2 million annually in 2017 and $227.2 million by 2026, growing at a CAGR of 29.5% (Hamad et al. 2019). The interest in green materials, sustainable and biodegradable conventional materials in emerging markets is driving the market. Increasing interest in innovations in paper, board and plastics, materials, food bundling, beauty care and personal cleaning, pharmaceuticals, biomedicine, paints, and coatings is driving the nanocellulose market.

Depending on the technology and method, the cost of CNC varies from $3 to $25 per gram, which is the same as the cost of carbon nanotubes. Considering that cellulose is the most abundant natural polymer and processing innovations have been created to remove cellulose from various sources compared to synthetic mercury fusion techniques for carbon nanotubes, CNC costs may become critical sooner or later. Facilitate. Similarly, another variable to consider when setting up CNC usage in different applications is extraction strategy. From now on, CNC is removed from cellulose hydrolysis by using various corrosive drugs. In order to obtain maintainable bio-based nanomaterials, new and ecosystem-friendly strategies should be developed (Trache et al. 2017).

The basic commitment of this part is to talk about the impact of CNC on nanohybrid materials. The advantages and disadvantages of basic processing methods for obtaining CNC-based nanohybrid materials are mainly studied. First, a short demo and dialogue on CNC characteristics is given. Important issues related to the production, handling, and dispersion of CNC-based nanohybrids are examined in the accompanying section. Influence of CNC on nanohybrid performance. CNC is divided as an extension of the program for the production of nanohybrid materials with further development characteristics focusing on design and biomedical applications. The natural effects associated with the use of CNC nanohybrids are also summarized. Finally, conclusions and future possibilities are presented and discussed.

5.2 Hybrid Manufacturing Technology

5.2.1 Solution Casting

Solution casting is the most widely used manufacturing strategy for obtaining pure or nanohybrid CNC materials (Marchessault et al. 1959). This technique is simple and feasible because it is likely done by scattering the CNC and enriching the nanoparticles into a liquid (usually water), which is then projected onto the Petri dish to drain the soluble substance (usually within 2 days). After drying at room temperature, the film can be stripped off the substrate. Achieving uniform scattering of nanohybrid partners in liquid arrangement becomes an essential element in obtaining highly dispersed CNC and homogeneous thin films. In this sense, sulfate CNCs are preferred because the electrostatic effect between nearby nanoparticles makes them very dispersed in the liquid arrangement (Fig. 5.1).

When CNCs transport nanoparticles, new chiral materials with practical overhaul properties are created (Revol et al. 1992). Typically, nanoparticles are mixed with CNC in water, and after proper scattering of CNC and nanoparticles in an ultrasonic step, they are projected onto the substrate to enable their vaporization-induced self-assembly, resulting in a strong nanohybrid film. Some important models are plasma-gold nanorods, quantum spots, carbon nanotubes, carbon swabs or unusual earth compounds (Revol et al. 1998).

In any case, the consolidation of such nanoparticles is largely limited to low loads (about 1–1.5 wt %), since their nature often interferes with the chiral nematic long-distance requirements of the CNC (Querejeta-Fernández et al. 2015). Advanced molecular stacking serves as the basis for the creation of multifunctional materials, because the more prominent the presence of responsive nanoparticles, the faster they react in the external booster. In this particular case, the preparation of

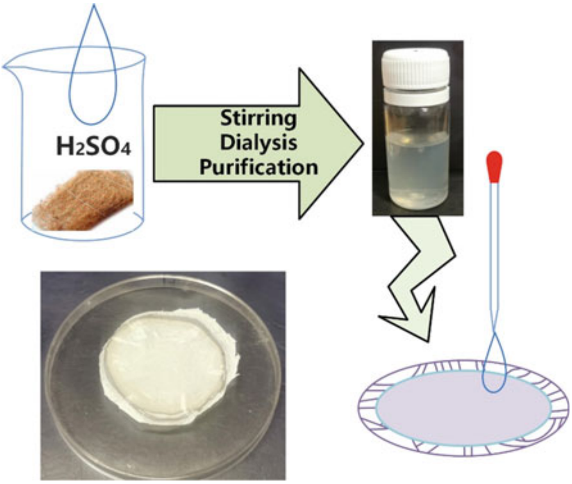

Fig. 5.1 Surface deposition of CNC on film

nanoparticles with suitable surface-useful aggregations to make them feasible with hydrophilic CNC is one of the main difficulties in this field (Lizundia et al. 2017a, b). Subsequently, hydrophobic nanoparticles can be surface-altered with amphiphilic stabilizers or parts such as carbonyl, hydroxyl and carboxylate aggregation can be transported.

5.2.2 Filtering Process

Using the traditional papermaking process, the aqueous CNC suspension can be quickly converted into a delicate film through a basic filtration process, including the extraction of a wet gel (called a gel/cake) and the subsequent disappearance of the water (Sehaqui et al. 2010). As the water in the gel/cake continues to disappear, a narrow power is generated that brings the CNC close to the point where the auxiliary spell is made, bringing full filaments with modulus of elasticity in the range of 5–10 GPa and rigid masses up to 70 MPa in ordinary films (Moon et al. 2011). While wet cakes can dry normally under the right conditions, several efforts have been made to reduce the time required for film making, including vacuum filtration of the CNC suspension, removal from channels, and subsequent broiler drying or autoclaving. While the cutting results are fascinating from a modern point of view, these projects, unfortunately, neglect the CNC's interesting ability to collect the self in very specific structures (Nan et al. 2017).

5.2.3 Layer by Layer Deposition

The layer-by-layer (LbL) strategy, first proposed by Iler in 1966, is a useful method for obtaining nanostructured faceted films with at least two species if they have an often attractive association (Iler 1966). While the idea was originally intended for countercharged polyelectrolytes, it has been extended to various frames, including CNC. In this production line, Martin et al. obtained diaspore trihydrate nanosheets with an absolute charge and a CNC facet film with an adverse charge. The results show that by fluctuating the ionic strength of the CNC suspension (via the expansion of monovalent salts), the layer thickness in the range of 13 to 70 nm can be controlled (Martin et al. 2017). Irradiation materials can also be obtained after LbL collection. For example, consolidation of colloidal SiO_2 and CNC, creating layers with alternative low and high refractive records, creates fragile films reflected at 550 nm (green).

With the LbL collection method, high-strength and very robust graphene oxide/CNC nanohybrids with layered morphology such as normal mother-of-pearl (7–15 nm per double layer) are obtained (Xiong et al. 2016). SA-LbL has also been extended to other nanohybrid frameworks, including multiwall carbon nanotubes (MWCNT)/CNC, aluminosilicate imogolite nanotubes/CNC, or zinc oxide/silver

(ZnO/Ag) (Trigueiro et al. 2014). Since these rely on static electricity, the robustness of faceted nanohybrids with communication as the main driving force may be slightly lower under brutal conditions, so the association between the two layers can be improved to bring various utilitarian aggregations into the CNC surface to provide a harder material (Li et al. 2018a, b).

5.2.4 Soft and Hard Templating

Given the combination of formats for CNC self-assembly, the LLC model was used to integrate mesoporous solids (porosity in the range of 2–50 nm) with large explicit surface areas and intermittent porosity (Kresge et al. 1992). CNC scattering is mixed with viable precursors, so self-aggregation delivers a nanostructured crossover material. At this point, one of the steps is specifically removed in order to obtain permeable material (Giese et al. 2015). According to this approach, controlled porosity can be brought into intermittent CNC structures, allowing CNC nanohybrids to help with applications such as drive support, enantioselective sensors, capacitor materials, or battery anodes. Using CNC as a template as a refined layout technique offers advantages, resulting in a particularly intriguing creation strategy compared to hard format strategies, as it allows good control of morphology, requires fewer manufacturing steps, and can be cultivated cost-effectively (Shopsowitz et al. 2011).

Taking into account the similarity of fast CNC autopolymerization and inorganic precursors during the sol–gel reaction, the mesoporous chiral nematic requirements obtained in CNC slimming films can also be transferred to many different materials by hard stencil strategy. For example, Shopowitz et al. used water scattering CNC as the chiral nematic stage. Free chiral nematic mesoporous carbon films with well-defined ranges up to 1465 m^2/g, meeting key instructions for mesoporous carbons with chiral requirements (CNC's EISA and TMOS, followed by pyrolysis at 900 °C followed by NaOH silica mapping in a nitrogen environment) (Shopsowitz et al. 2011). In addition, chiral nematic CNC/silica films can be calcined in air at 540 °C, leaving a silica gel film with a large surface area. The mesoporous chirality of CNC can also be transferred to various materials by unloaded mesoporous films of polymers formed by silicon dioxide, metal/metal oxides or fluorescence (Shopsowitz et al. 2010).

5.2.5 Nanoparticle Growth onto CNC

Due to the high surface energy of the nanoparticles, the overall inclination of the nanoparticles after mixing can be overwhelmed by turning into inorganic nanoparticles directly on the CNC surface (Kaushik and Moores 2016). With CNC as a strong format, nanoparticles can be developed by aqueous or decreasing techniques. For example, ZnO nanoparticles are developed on a water-scattering CNC covered with an elongated melamine–formaldehyde layer (Awan et al. 2018). At 100 °C, the water hydration of zinc acetate derivatives is absent and NaOH and CNC produce round ZnO nanoparticles of 20–25 nm. Heat CNC, polyethylene glycol and chlorine corrosively at 80 °C for 1 h (Yan et al. 2016).

5.2.6 Sol–Gel Process

The sol–gel process involves the conversion of a "sol" (colloidal arrangement) into a "gel"-like frame with a strong and liquid phase. The morphology of these two stages can range from discrete particles to uninterrupted polymer tissue. When using water as the liquid stage (the most famous case of CNC), the sol–gel process creates a hydrogel, a three-layer, deeply penetrating hydrophilic fabric suitable for absorbing large amounts of water, thus keeping the tissue structure flawless. Using the remote-controlled chiral nematic requirements of CNCs, an improvement was made in response to photonic hydrogels in 2013, showing large color changes in the light of pH, solubility or temperature changes (Kelly et al. 2013). The chiral nematic phase is maintained by connecting the CNC to a nonionic hydrogel predecessor (acrylamide, acrylic corrosive, 2-hydroxyethyl methacrylate, macrogol methacrylate or N-isopropylacrylamide as monomer). Under ultraviolet light, the self-collected design is blocked. In addition, Hiratani et al. developed CNC hydrogels for ionic strength and strain detection. An anisotropic structure with critical birefringence was obtained by mechanical shear (Hiratani et al. 2018).

Although these models require CNC to facilitate the gelation of polymers, there is basically less work to manage pure CNC hydrogel improvements (there is no polymer stage, only CNC and water) (Ureña-Benavides et al. 2011). The production of pure CNC hydrogels has been cultivated in many ways. In order to create a consistent organization of the detected poles from the nano to the macro scale, the simplest approach is to increase the CNC focus in the dispersion by more than 14 vol % (Heath and Thielemans 2010). Low-power ultrasound also creates the gel 3D fabric of a CNC because hydrogen remains bonded between adjacent CNCs. A finer way to deal with acquired hydrogels is to control static aversion between adjacent CNCs. Drops, which can weaken the dispersion of the charge balance. In this way, bonding the salts protects the carbonized electrical double layers, enhances their zeta potential, and reduces the electrostatic aversion between the CNC along these lines. After the salt expands, the charm becomes stronger, resulting in CNC collection and the resulting hydrogel (Dong and Gray 1997).

A green, immediate, and effective CNC hydrogel manufacturing technology was recently announced that uses water treatment to advance the desulfurization of CNC surfaces, reducing their dispersibility and finally favoring their hydrogel arrangement. The results show that adjustable mechanical properties can be achieved by controlling the water temperature, response time, CNC focus, and pH. Similar meetings also show the production of CO_2-switchable CNC hydrogels, in which the gelation is due to the expansion of ionic strength by imidazole protonation (Bertsch et al. 2017; Oechsle et al. 2018).

5.2.7 Oven-Drying, Freeze-Drying, and Supercritical-Drying

The fluid circuit of the hydrogel can be removed by various techniques to obtain primary strong materials with deep penetration (close to 100%) and ultra-low thickness nanostructures (4–500 mg/cm^3) with a large exposed surface area (100–1000 m^2/g). Kistler first announced this permeable construction, called aerogels, in 1931 and can be obtained by various drying processes, of which freeze-drying and supercritical drying are the most popular methods (Pierre and Pajonk 2002). Previously, various aerogels made of carbon, silicon dioxide, alumina, and tin oxide were considered. The production of deep penetration CNC designs can help to improve specific adsorption properties, thermal and electrical protection properties, acoustic materials for attenuation limits and as templates/beams for various materials, thus opening new doors (Baetens et al. 2011).

The soluble evacuation step essentially influences the morphology and properties of subsequent aerogels and remains an important test for their formation. In fact, the disappearance of fluids in osmotic systems stresses fine stretches, leading to pore deformities, aerogel shrinkage, and surprising primary decompositions and fractures (Scherer 1986). Ice sublimation avoids the development of liquid-smoke interfaces and limits the presence of fine stresses that damage the structure. For example, the first hydrogel structures can be protected by submerging the fast-freezing solutes with liquid nitrogen, while slow freezing is often used as a way to create designs. In this strategy, known as the “ice template” (IT), the distribution of the propagation phases is carried out by the development of ice gemstones (Deville 2010). The construction of ice gemstones can be limited by directed warm slopes to create honeycomb 2D shapes that are compatible with the freezing process and offer interesting anisotropic mechanical properties (Munier et al. 2016).

5.2.8 Electrospinning

The innovation of electrospinning is gaining importance in the production of practical materials, as it is used to produce a layer of nano- and microfibers without interruption (Reneker and Yarin 2008). After using the electric field, the response containing the given material (broken, scattered or liquid) is removed from the spinnerets and stored with the help of electrostatic communication on the authority to provide a fiber pad of electrospinning (Ahmed et al. 2015). Boundaries such as fixed permutation, resolvable decision, applied stress, distance between tip and authority, and relative blockage fundamentally influence the morphology of the subsequent material. During electrospinning, the polar CNC adapts to the main fiber pivot point, resulting in an anisotropic material. Unfortunately, due to the difficulty of producing CNC in successive stages, CNC was mainly used as a support for the gradual relief of polymer fiber felts, and no work was announced on the development of pure CNC electrospinning. However, given the excellent scattering of CNC in water, we recognize the extraordinary potential of electrospinning innovations in creating novel CNC nanohybrids with tendon morphology (Zhou et al. 2011).

5.3 Nanohybrid Types

5.3.1 CNC Structural, Chemical and Physical Properties

When considering the synthetic and practical properties of CNCs, it should be noted that their properties depend in particular on the strategy used for separation. CNC is removed from cellulose by controlled hydrolysis, in which less coordinated (blurred) areas endure the cleavage of their glycosidic bonds, leaving areas with safer, deep-translucent areas. In 2009, an enzymatic hydrolysis process was carried out to obtain CNCs, and most of their production was done after corrosive hydrolysis of solids (Filson et al. 2009). Typically, cellulose is immersed at 45° C for 30 min in a sulfuric acid corrosive (H_2SO_4) (64 wt.%) liquid arrangement and stably mixed, but other acids such as phosphoric acid, nitric acid, hydrobromic acid or hydrochloric acid can also be corrosive. After additional mechanical treatment by ultrasound, a water-scattering CNC was obtained (Lizundia et al. 2016).

CNC is a needle-like translucent nanoparticle of β-1,4 bonded anhydrous D-glucose units with widths of 3–20 nm, while its length can vary from 150 to 2000 nm depending on the cellulose source and mixing conditions, although usually a quality of 100–250 nm is obtained. CNCs typically have a degree of polymerization (DS or glucose units) of 500–15,000 and, due to their anisotropic properties, a specific surface area in the range of 150 and 250 m^2/g (Beck-Candanedo et al. 2005; Habibi et al. 2010).

5.3.2 CNC/Metal Oxide Nanohybrids

Oxide metal nanoparticles (NPs) such as titanium dioxide (TiO_2), zinc oxide (ZnO), copper oxide (CuO and Cu_2O), silicon dioxide (SiO_2) and iron oxides (Fe_2O_3 and Fe_3O_4) require their versatile, modulating and multifunctional properties, which are fundamental for relatively long-term technological applications, including propulsion, safety precautions, optoelectronics, attractive nanocomposites, luminescent materials, drug delivery frameworks, sensors, antimicrobial materials, imaging (Boury and Plumejeau 2015; Goikuria et al. 2017).

5.3.3 CNC/Carbonaceous Nanomaterials

Carbon nanostructures can be grouped according to their perspective ratio and nanometer size, using 1D as nanotubes or nanofibers, 2D as graphene nanosheets or graphene oxide, and 3D as fullerenes or carbon-dark nanoparticles (Iijima and Ichihashi 1993). Carbon nanotubes (CNTs) are formed by moving the graphene layer upwards in a circular and hollow manner. They are sequenced into single-walled carbon nanotubes (SWCNTs), double-layer carbon nanotubes (DWCNTs), and multi-walled carbon nanotubes (MWCNTs) as indicated by the number of layers, creating containers of different sizes and dimensions, ranging in length from about a few nanometers to millimeters, by changing interactions and boundaries.

Graphene is a two-layer carbon material consisting of a single sheet of graphite. The monolayer of carbon particles is organized in a hexagonal cross-section so that a thickness of 0.35 nm is assumed. Andre Geim and Kostya Novoselov received the Nobel Prize in Physics in 2010 for “leading the study of the double-layer material graphene” (Geim and Novoselov 2010). It is extremely conductive and robust, with a modulus of elasticity of 1 TPa and an extreme strength of 130 GPa. In fact, it is a material with excellent mechanical properties, even compared to carbon nanotubes. Fullerenes are an allotrope of carbon that is round (Zhu et al. 2010).

All carbon nanostructures have very strange electrical properties, and they are mixed with other nanomaterials to transfer this effect to the membrane material. Carbon nanostructures enable improved low-conductive layers of carbon CNC nanohybrids. Most conductive hybrids use carbon nanotubes or graphene, two new materials that often have amazing properties. Carbon black (CB) is additionally used due to its innate conductivity over a large surface area (Meng and Manas-Zloczower 2015).

5.3.4 CNC/Luminescent Nanoparticles

The resulting combination of brilliant materials and luminescence was sought as a tempting way to plan new optical devices. Quantum spots (QDs) are small semiconductors that have tunable and elongated emission spectra induced by quantum repulsion, which is particularly useful for detection and light exchange (Alivisatos 1996). Quantum swabs are passivated with a suitable amphiphilic stabilizer to keep them away from their accumulation and ensure the development of chiral nematic phases. The photoluminescence (PL) outflow power of the film corresponds to the QD stack, which shows green to red light when irradiated with ultraviolet light (emission frequency of 490–680 nm) (Bruchez et al. 1998).

Carbon Pats (CDs) without metallic photoluminescent nanomaterials are attracting more and more attention due to their lowest cost, PL stability, and intrinsic physicochemical properties. Contrary to what is commonly found in many semiconductor nanoparticles, the photoluminescence of CDs often relies on precursor materials used for their combination, making them useful in the fabrication of materials for detection, catalysis or design (Baker and Baker 2010).

5.4 Applications of CNC Nanohybrids

The study of CNC inorganic/natural nanohybrids is a rapidly developing interdisciplinary field in materials science and design that provides valuable properties for two parts that cannot be observed alone. In this sense, there are some licenses, progressive and commercial projects (George and Sabapathi 2015). Cellulose nanocrystals have been read and evaluated for a variety of applications, particularly in paper, polymers, plastics, chiral layout, flocculants, aerogels, hydrogels, drug delivery, detection, catalysis, high energy, and biomedicine (Fig. 5.2).

5.4.1 Energy Applications: Supercapacitors, Solar Cells, Batteries

Cellulose nanocrystals offer the benefits of high strength and adaptability for some advanced applications, providing capacitor gadgets that are less expensive than battery-powered batteries, with higher power thicknesses and faster charging capabilities (Dhar et al. 2018). Fast chargers can save critical energy because they accumulate energy when slowing down and provide energy as the speed increases. While conventional supercapacitors can actually damage the climate during assembly and dismantling, cellulose-based electroactive nanohybrids can be calibrated to make them more adaptable on the nanoscale (Pérez-Madrigal et al. 2016).

As is shown in Fig. 5.3, CNC can be used as (I) lightweight mechanical substrates with adaptability and unity to electrochemical and electroactive materials; (ii) conventional layout covered with other electrochemical dynamic anode materials or converted into other carbons by pyrolysis; (iii) the stomach between the cathodes of the supercapacitor; (iv) Materials that promote electrolyte retention, supplied as electrolytes or even as components of the actual electrolyte (Borghei et al. 2018).

5.4.2 Environmental Remediation Application

Practical cellulose-based materials are often used for separation, especially in the field of water treatment (Fig. 5.4). Biopolymers such as chitosan, chitin, and lignin are

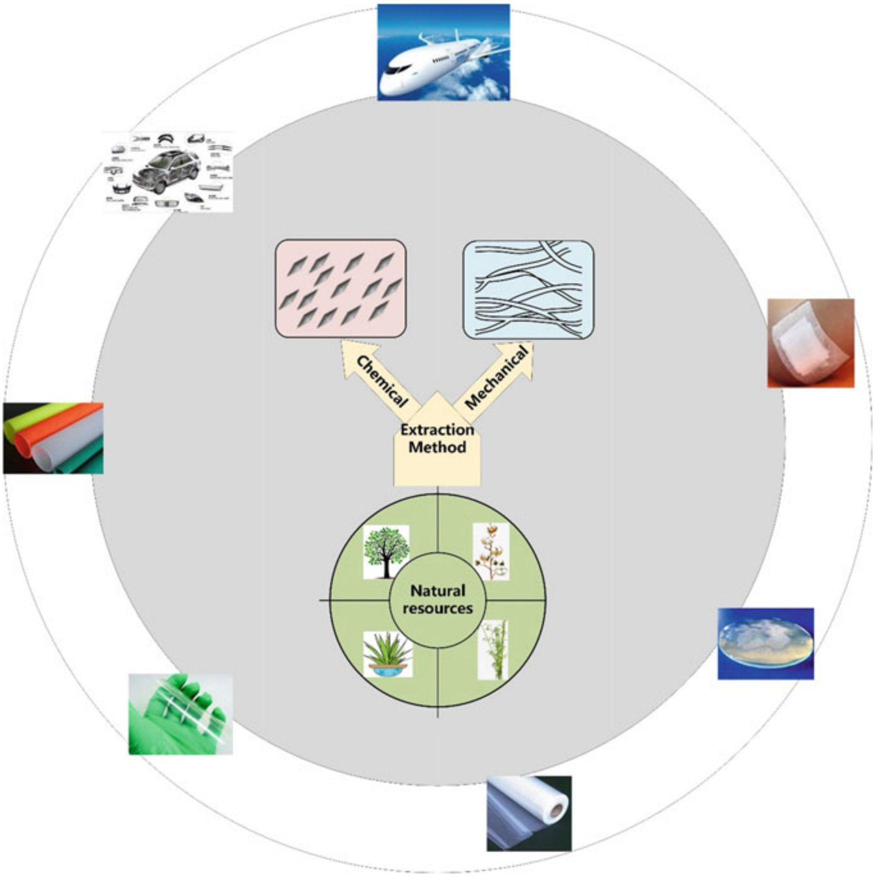

Fig. 5.2 Extraction and application of nanocellulose from lignin biomasses

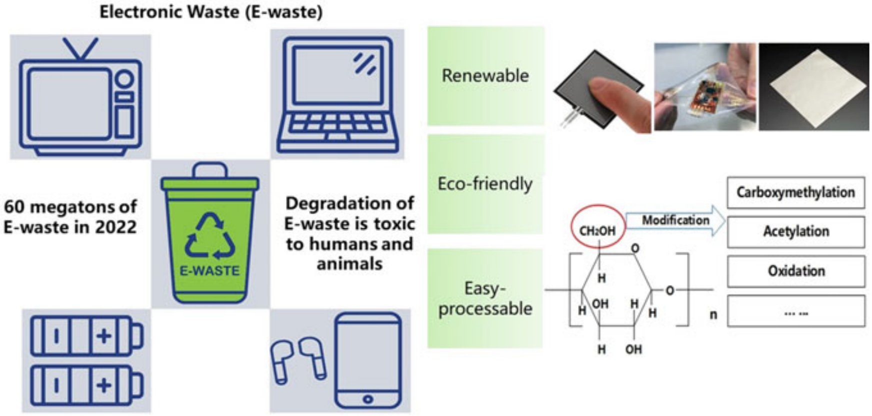

Fig. 5.3 Application of cellulose-based materials in electronic components

known to adsorb heavy metal particles from liquid arrangements. Practical cellulose-based materials offer excellent adsorption capacity for water treatment applications (Li et al. 2018a, b).

Nanotechnology is believed to be able to limit costs and further increase productivity in predicting, treating, and remediating pollution. Two applications of cellulose nanostructures in this field have aroused interest, in particular as dynamic adsorption materials for impurities and stabilizers for other dynamic particles (Carpenter et al. 2015).

Cellulose nanomaterials have an effectively functionalized surface that can limit the performance of nanocellulose impurities due to the bonding of material parts.

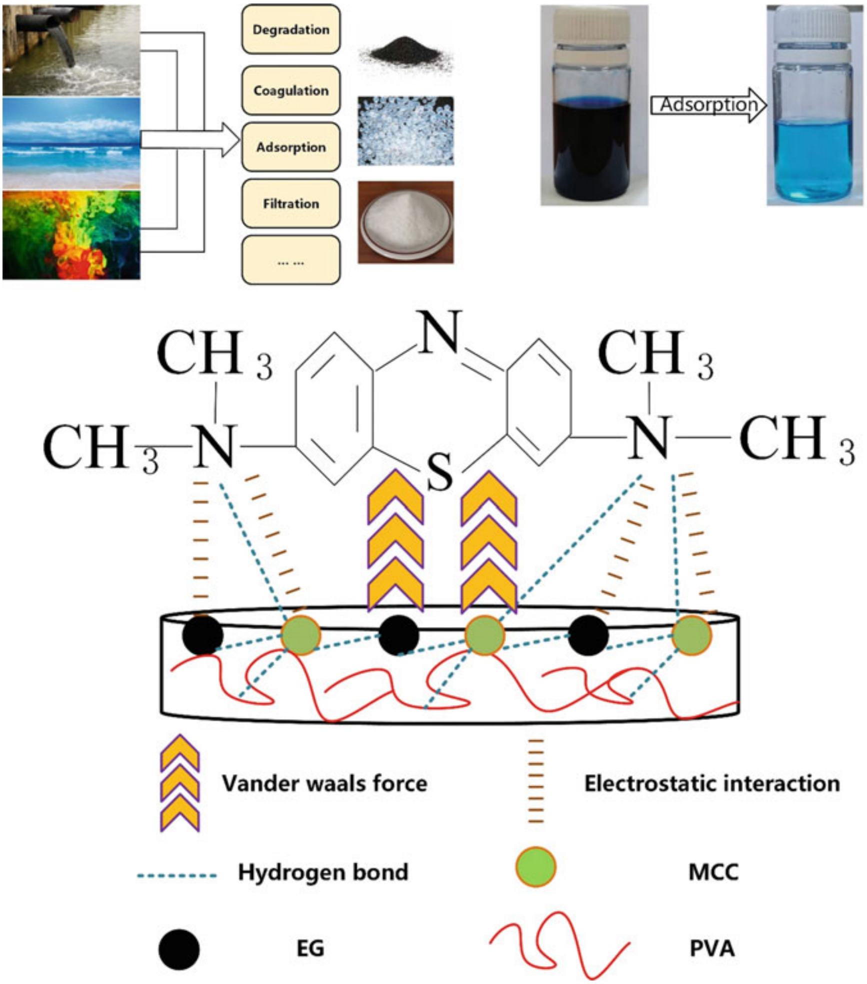

Fig. 5.4 Schematic diagram of the mechanism of physical adsorption of waste dyes on cellulose-based materials

Carboxylation is the technology that focuses most on increasing the adsorption limit. Yu et al. found that attaching succinic acid blasting to CNC significantly limits the productivity of Pb^{2+} (Yu et al. 2013).

5.4.3 *Catalysis*

Catalysis plays an important role in both industry and science, as it provides an effective platform for triggering various chemical reactions (Pan and Wang 2012). For example, catalysis can be used to produce materials such as polymers or for environmental remediation purposes by breaking down environmentally harmful molecules. The decomposition of organic pollutants in water offers the opportunity to reduce the widespread accumulation of pollutants in reservoirs (i.e., antibiotics, insecticides, etc.), a global challenge of the twenty-first century (Salas et al. 2014).

Since catalytic reactions often occur in the outer layers of valuable/non-valuable metallic materials, nanoscale particles are preferred because they have a larger explicit surface area and an increased number of reaction sites per unit mass (Eisa et al. 2018). Unfortunately, many people use nanoparticles to disperse synergistic reactions in liquid media, reducing their pleasantly explicit surfaces, and it is difficult to recover the propulsion particles once their capabilities are complete. To solve these difficulties, reactant nanoparticles can be fixed to various auxiliary materials such as cellulose, where the surface hydroxyl bundle provides great adhesion between the particles and the nanocellulose. Since this field has been considered recently, new light has been gained in heterogeneous catalytic processes with CNC as carrier material (Wu et al. 2013).

5.5 Conclusion

This paper provides an overview of the latest major improvements in the use of CNC in the field of nano-based nanocontamination of cellulose, including CNC design, inorganic or natural nanoparticle mixtures for the arrangement of nanopollutants and their applications. Due to its adaptability, CNC is one of the most interesting nature-based nanomaterials with a number of remarkable properties, including innate repeatability, biodegradability, business accessibility, adaptability, printability, low thickness, high porosity, optical directness, and superior mechanical, thermal, and physico-chemical properties.

Although some progress has been made in the production, characterization, and use of CNC or CNC-based nano-grown materials, there are still some problems that need to be solved from a logical and mechanical point of view.

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