Review

Nanocellulose-Based Materials for Water Treatment: Adsorption, Photocatalytic Degradation, Disinfection, Antifouling, and Nanofiltration

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Abstract: Nanocelluloses are promising bio-nano-materials for use as water treatment materials in environmental protection and remediation. Over the past decades, they have been integrated via novel nanotechnology approaches for water treatment processes. This review aims at giving an overview of nanocellulose requirements concerning emerging nanotechnologies of water treatment and purification, i.e., adsorption, absorption, flocculation, photocatalytic degradation, disinfection, antifouling, ultrafiltration, nanofiltration, and reverse osmosis. Firstly, the nanocellulose synthesis methods (mechanical, physical, chemical, and biological), unique properties (sizes, geometries, and surface chemistry) were presented and their use for capturing and removal of wastewater pollutants was explained. Secondly, different chemical modification approaches (surface functionalization) for enhancing the surface chemistry of the nanocellulose for enabling the effective removal of specific pollutants (suspended particles, microorganisms, hazardous metals ions, organic dyes, drugs, pesticides, fertilizers, and oils) were highlighted. Thirdly, new fabrication approaches (solution casting, thermal treatment, electrospinning, 3D printing) that integrated nanocelluloses (spherical nanoparticles, nanowhiskers, nanofibers) to produce water treatment materials (individual composite nanoparticles, hydrogels, aerogels, sponges, membranes, and nanopapers) were covered. Finally, the major challenges and future perspectives concerning the applications of nanocellulose based materials in water treatment and purification were highlighted.

Keywords: nanoparticles; nanocrystals; nanowhiskers; nanofibers; hydrogels; bacterial cellulose; surface functionalization; membranes filtration

1. Introduction

Nowadays, treatment and purification of sanitation and industrial water are very critical to the survival of people and the planet. About 97.5% of the total available water on earth is salty which is not suitable for use [1]. The remaining water is 2.5% of fresh water, only ~1%, of this fresh water is available for human consumption. Of the total wastewaters
produced from human activities, 10–20% of all the wastewater is treated and reused. Therefore, billions of people around the world lack access to safe drinking water and sanitation [1]. The freshwater is contaminated due to human activities and consequently is enriched in foreign and potentially dangerous species. These pollutants can be classified into four categories: microorganisms (virus, bacteria, fungi), inorganic compounds (heavy metals, and radioactive materials), organic compounds (soaps, drugs, fertilizers, pesticides, and oils) [2]. They are all harmful to human health and the environment, induce changes in natural aqueous habitats and organisms, and affect the water quality and biodiversity. However, due to the complexity of wastewater types and the huge diversity of pollutants, specific water treatment strategies have been developed and experts are required to select the most appropriate filtering techniques and membrane materials [3].

Nanocelluloses have emerged as an alternative to conventional wastewater treatment materials. It can be produced from different sources, particularly soft and hardwood species, phloem fibers (flax, hemp, jute, ramie), grasses (bagasse, bamboo), and also bacteria, fungi, algae, and marine invertebrates [4–7]. Due to its intrinsic features, nanocelluloses based wastewater treatment materials are recommended candidates for industrial water treatment systems [8]. They can serve this goal by providing highly efficient materials for wastewater treatment owing to their high aspect ratio, surface area, surface charge, and mechanical strength. The performance of nanocelluloses based wastewater treatment materials (e.g., adsorbents, membranes, flocculants, photocatalysts, and disinfectants) has to fulfill diverse functions. However, nanocelluloses functions must be provided with an optimum balance between porosity, high permeability, selective binding, durability, and specific filtration mechanisms (i.e., size exclusion, ion exchange, adsorption). Moreover, the size, morphology, and surface chemistry of nanocelluloses need to be controlled for simultaneously tuning the affinity towards specific pollutants and avoiding microbial growth and fouling [3,9].

This review provides an overview of types, classifications, and unique properties of nanocelluloses as well as the recent methods for nanocellulose synthesis and control of their sizes, aspect ratio of a geometric shape, and pathways to rationalize their surface properties for filtration via size exclusion, absorption/adsorption, flocculation, photocatalytic degradation, disinfection, and antifouling. Intrinsic adsorption properties of nanocellulose [10], and filtration membranes [11–13] are highlighted in previous reviews for wastewater treatment [14]. In contrast with previously published reviews, this multidisciplinary article offers an updated and critical assessment of recent findings on nanocellulose and cellulose nanocomposite research with a focus on anti-microbial activity, photocatalytic degradation, disinfection, and antifouling, ultrafiltration, nanofiltration, and reverse osmosis to water engineers and experts in chemistry or materials science. The review discusses different chemical modification approaches for enhancing the surface chemistry of the nanocellulose based materials towards the effective removal of specific pollutants such as hazardous metal ions, organic dyes, drugs, pesticides, fertilizers, and oils.

2. Production, Morphologies, and Unique Properties of Nanocelluloses

The industrial-scale production and market of nanocellulose are rapidly expanding worldwide as they are environmentally friendly, non-toxic, sustainable, low-cost, and highly efficient materials for a wide variety of applications, including water treatment. At present, different forms of nanocellulose are available on the market, which is forecasted to achieve USD 783 Million by 2025 according to Markets and Markets. The global market size of nanocelluloses is expected to grow at 21.4% of a compound annual growth rate from 2020 to 2026 [5]. The main sources of nanocellulose are plants, whereas bacteria and tunicates (sea animals) are currently less used to obtain nanocelluloses [15]. To date, researchers have developed several nanofabrication techniques for the production of nanocelluloses: (1) Mechanical methods such as grinding, homogenization, refining, aqueous counter collision, cryo-crushing are normally used to disrupt the cellulose microfibers from plant materials down to submicron or nanoscale dimensions [16]. Typically, the high energy
input and the microfibers suspensions with extremely high viscosity must be optimized to high yield nanocelluloses at low cost. (2) physical techniques such as steam explosion, wet spinning, dry spinning, melt spinning, or electrospinning have been used to produce electrospun cellulose acetate nanofiber (100 to 1000 nm in width and over 1 µm in length) [16,17]. Cellulose aerogels (CAG) have been fabricated by the dissolution of nanocelluloses in a favorable solvent, followed by gelation, cross-linking, solvent exchange, and drying. (3) Chemical treatment of lignocellulose materials including alkali treatment, acid hydrolysis, sulfonation, and ionic liquid approaches with selected chemicals used to provide nanocelluloses with specific functionalities and high purity [15,18,19]. (4) biological treatment such as enzymatic hydrolysis has been applied before mechanical cellulose fragmentation to reduce chemical waste and energy consumption [5,20]. Figure 1 and Table 1 summarizes the classification of the nanocelluloses according to their length, diameter, aspect ratio, and composition.

**Figure 1.** Electron microscope micrographs of different types of cellulose nanomaterials: (a) cellulose microfibers (CMFs) [21], © Springer Nature, 2021; (b) Cellulose fibers (CNFs) [22], © Hindawi, 2017; (c) Spherical cellulose nanoparticles (SCNPs) [23], © Elsevier, 2007; (d) Cellulose nanocrystals (CNCs) [24], © MDPI, 2008; (e) cellulose nanofibers (CNFs) [25], © Royal Society of Chemistry, 2009; (f) bacterial cellulose (BC); (g) cellulose nanofibers assembled into a material stronger than spider silk [26], © American Chemical Society, 2018; (h) Electrospun cellulose nanofibers (CNFs) [27], © Elsevier, 2014; (i) Cellulose aerogels [28], © MDPI, 2018.
Table 1. Classification of nanocellulose materials according to their morphology, origin, and mean size.

| Type                                         | Origin                                      | Mean Size                                           | Ref.  |
|----------------------------------------------|---------------------------------------------|-----------------------------------------------------|-------|
| Cellulose Microfibers (CMF)                  | Wood, cotton, hemp, flax, straw            | Several microns in diameter and hundred microns in length | [29]  |
| Cellulose nanofiber (CNF)                    | Wood, cotton, hemp, flax, straw            | Few microns in diameter with finely attached nanofibers; several to hundred microns in length | [30]  |
| Spherical cellulose nanoparticles (SCNPs)    | Wood, cotton, hemp, flax, straw            | Diameter: 10–100 nm                                 | [31]  |
| Cellulose nanocrystals (CNC)                 | Wood, cotton, hemp, flax, wheat straw, rice straw, mulberry bark, ramie, Avicel®, tunicin, algae | Width: 5–70 nm Length: 100–250 nm (from plants); 100 nm to several micrometers (from the cellulose of tunicates, algae, and bacteria) | [6]   |
| Bacterial nanocellulose (BC)                 | Low molecular weight sugars and alcohols    | Width: 5–70 nm Length: several micrometers          | [32]  |
| Cellulose nanoyarns (CNY)                    | Cellulose derivatives                       | Diameter: 100 to 1000 nm Length: >1 µm              | [6]   |

Characteristics of nanocelluloses (size, morphology, aspect ratio, surface charge) can be modulated by selecting specific raw materials, fabrication techniques, and processing parameters. The favorable characteristics of nanocellulose for employment in wastewater treatment are justified through their unique physicochemical properties (Table 2), i.e., high surface area, high aspect ratio, high mechanical stiffness, high crystalline degree, susceptible to surface functionalization, stability in water, and high surface tension. Besides these characteristics, enhanced rheological properties, alignment, and orientation, biodegradability, biocompatibility, renewable and abundancy, chemical inertness, low coefficient of thermal expansion, low density and dimensional stability are particularly interesting for water treatment applications. The nanocelluloses presents ultra-fine pore size, high surface area, and high amount of hydroxyl group [33]. The hydroxyl groups are negatively charged and exhibit electrostatic attraction toward cationic wastewater pollutants, such as heavy metals and cationic dyes, but they can also be chemically modified to improve the nanocellulose affinity for anionic and non-ionic pollutants. Interactions of water pollutants with nanocellulose surface can be further tuned through surface grafting (carboxylic, sulfate, phosphate, ester, salcilate groups) [17] or decoration with inorganic nanomaterials (metals, metal oxides, clay [34], zeolites [35], carbon nanomaterials (graphene, carbon nanotubes, active carbon [36] or polymers (ion exchange resins [37] chitosan [38–40] alginate [41], and lignin [42]. However, certain current drawbacks hinder the further expansion of the nanocellulose utilization as mainly related to their processing conditions:

1. High dispersion stability of nanocelluloses in aqueous solutions can be achieved through sulfuric acid hydrolysis introducing negatively charged sulfate groups. However, this high dispersion stability of individual nanocellulose particles makes their separation from the water system difficult and necessitates the addition of salt or pH alteration to recover them after the water treatment process.

2. Dispersion of nanocellulose in hydrophobic polymer matrices (membranes) remains a critical issue. However, the dispersion of nanocelluloses in polymer blends for sustainable wastewater treatment applications can be achieved by surface grafting of nanocelluloses with low molecular weight polymers. Solution-casting is the most important method for preparing nanocellulose-polymer composite membranes, which, however, remains difficult for large-scale application.

3. Production of nanocellulose from plant sources is generally based on multi-step, top-down techniques that include physical (e.g., refining, mechanical grinding, ultrasonic grinding, thermal treatment), chemical (e.g., acid hydrolysis, alkali treatment, and...
chemical modification), biological (e.g., enzymatic hydrolysis and production of cellulose nanofibers from bacteria), and hybrid methods [43,44]. High water and energy consumption and yield are the main challenges in the preparation process, along with by-product toxicity [4]. For example, acid wastewater is typically generated from the washing process for neutralizing the pH value of the nanocellulose suspension [45].

Table 2. Relation between the physicochemical features of nanocelluloses and their adsorption capacity for hazardous pollutants in the aqueous environment.

| Nanocellulose Features          | Effect on Adsorption                                                                 | Reference   |
|---------------------------------|--------------------------------------------------------------------------------------|-------------|
| High surface area               | Increases the specific surface area from micro- to nano-size, thus enhancing the nanocellulose adsorption capacity. The mixed aerogel (ratio of 1:3 CNC/CNF) can provide a higher specific surface area than pure CNC or CNF. | [43,45]     |
| High aspect ratio               | Aspect ratio of CNC (10–80) is generally smaller than that of CNF (up to 80–500), depending on nanocellulose sources and the treatment process. Favors the set-up of percolated CNCs and entangled CNF networks held by strong hydrogen bonding, thus enhancing the adsorbent mechanical strength | [45]        |
| High mechanical stiffness       | CNC and CNF high mechanical stiffness (modulus), ~130 and ~70 GPa, respectively, increases the adsorbent material stiffness and cohesion. High crystalline forms (CNCs and CNFs) are transparent, and gas-impermeable with a very high tensile strength up to 8 times that of steel. | [6,46]      |
| High crystalline degree         | Nanocellulose high crystallinity degree (60–80%) enhances the adsorbent chemical resistance and reduces cellulose solubility even in high polar solvents. | [47]        |
| Susceptible to surface functionalization | Hydroxyl group functionalization (oxidation, esterification, etherification, radical grafting, and silylation) increases the nanocellulose adsorption capacity | [48,49]    |
| Stability in water              | Reduces biofouling of nanocellulose-based adsorbents. The surface of cellulose-based water treatment materials is negatively charged due to the high concentration of hydroxyl and carboxylate groups, resulting in higher electrostatic repulsive forces between the surface layer and most model foulant. | [50]        |
| High surface tension            | High surface tension (nanocellulose surface energy is ~60 m J m⁻²) of nanocellulose-based adsorbents by water improve the wetting characteristics and reduce the bio-fouling | [51]        |

3. Adsorbents for Hazardous Metal Removal

Hazardous metal ions (e.g., Ni²⁺, Ag⁺, Cd²⁺, Cu²⁺, Pd²⁺, Hg²⁺, U⁶⁺) originating from battery manufacturing, petroleum refining, metal plating drainage, mining activities, paint manufacturing, and photographic products, are abundantly released in the environment [51–54]. The pollution of agricultural soil causes the wide distribution of toxic heavy metals in the environment, and this affects the microorganisms and plants’ growth. Exposure to heavy metals (oral ingestion, inhalation, and dermal exposure into humans) can cause damage to the lungs, liver, kidneys, and other organs. Radioactive and heavy metal ions have been found to interact with cell components such as DNA and nuclear proteins, causing DNA damage. Prolonged exposure to toxic heavy metals causes cancers (i.e., prostate, stomach, kidney, urinary system, and bones) and Alzheimer’s disease [55]. From this perspective, it is necessary to develop green treatment strategies to remove hazardous heavy metals from the industrial water system [52].

To date, several methods (chemical precipitation, adsorption, reverse osmosis, solvent extraction, and electrochemical treatment) have been employed to remove radioactive and
heavy metals from contaminated water [53]. Adsorption of hazardous (radioactive and heavy) metal ions is considered as one of the suitable water treatment methods due to due to its high efficiency, low cost, and ease of operation. Numerous studies reported that the nanosorbents remove radioactive and heavy metals from wastewater, e.g., carbon tube, graphene oxide, polymeric, zeolites, metal and metal oxides nanosorbents [54]. For using nanocellulose-based adsorbents, ion exchange and chemical-complexation are the main two mechanisms concerned for the uptake of heavy metals (Figure 2). The ion-exchange mechanism involves the adsorption of hazardous metal ions (M$^{n+}$) takes the place of other ions (K$^+$, Na$^+$, H$^+$) already associated with the nanocellulose surface (Figure 2a). In chemical complexation, the carboxyl (-COO$^-$) and hydroxyl (-OH) groups of the nanocelluloses have specific site interactions with particular hazardous metal ions (M$^{n+}$) (Figure 2b). The maximum adsorption capacity of nanocelluloses is limited by their surface area, functionality, and stoichiometry rules which cannot exceed half the content of surface ionic sites. For this reason, increasing surface area and surface functionalization is necessary to increase or introduce more complexing sites on which the hazardous metal ions can be adsorbed. Most work related to the usefulness of nanocellulose as an adsorbent for hazardous metal ions involved CNF [55–59], while limited works have been reported on CNCs and BNCs. The high surface area and nature of the functional groups on nanocelluloses drive their sorption efficiency. Table 3 lists the various nanocelluloses used as adsorbents to eliminate hazardous metal ions from contaminated wastewater.

![Figure 2](image_url)

**Figure 2.** Heavy metal removal mechanism from water system using nanocelluloses: (a) Ion exchange mechanism which involves the adsorption of hazardous metal ions (M$^{n+}$) takes the place of other ions (K$^+$, Na$^+$, H$^+$) already associated with the nanocellulose surface; (b) chemical complexation mechanism in which the carboxyl (-COO$^-$) and hydroxyl (-OH) groups of the nanocelluloses have specific site interactions with particular hazardous metal ions.
Table 3. Nanocellulose materials and their surface modifications are exploited as adsorbents for heavy metal pollutant removal from wastewater. BC—bacterial cellulose.

| Nanocellulose | Surface Modification | Heavy Metal | pH | Removal Efficiency (mg/g) | Langmuir | Freundlich |
|---------------|----------------------|-------------|----|---------------------------|----------|------------|
|               |                      |             |    |                           | Q<sub>max</sub> (mg·g<sup>-1</sup>) | K<sub>L</sub> (L·mg<sup>-1</sup>) | R<sup>2</sup> | K<sub>F</sub> | 1/n | R<sup>2</sup> |
|               |                      | Ag<sup>+</sup> | 6.39 | 34.35 | 1.9 | 0.09 | 0.996 | - | - | - | [60] |
|               |                      |            | 3.5–4.5 | 56 | - | - | - | - | - | - | [61] |
| Sulfate (-SO<sub>3</sub>−) |                      | Cd<sup>2+</sup> | 6.0 | 8 | 1.9 | 0.09 | 0.996 | - | - | - | [62] |
|               |                      |            | 6.5 | 9.7 | 11.2 | 0.63 | 0.95 | 5.75 | 4.71 | 0.92 | [63] |
|               |                      | Cu<sup>2+</sup> | 3.5–4.5 | 19 | - | - | - | - | - | - | [61] |
|               |                      |            | 6 | 0.59 | - | - | - | - | - | - | [64] |
| Phosphorylation (-PO<sub>3</sub>−) |                      | Pb<sup>2+</sup> | 5.5 | 47 | 27.9 | 0.04 | 0.973 | - | - | - | [62] |
|               |                      |            | 6.5 | 9.42 | 10.20 | 1.81 | 0.98 | 7.06 | 6.89 | 0.94 | [63] |
| Succination (-COO−) |                      | Fe<sup>3+</sup> | 3.5–4.5 | 6.3 | - | - | - | - | - | - | [61] |
|               |                      | Cr<sup>3+</sup> | 6.5 | 1.6 | - | - | - | - | - | - | [65] |
| Cellulose nanocrystals |                      | Cr<sup>4+</sup> | 2.5 | 0.6 | - | - | - | - | - | - | [65] |
|               | Sodium salt succination (-COO− Na<sup>+</sup>) | Ni<sup>2+</sup> | 6.5 | 8.55 | 11.23 | 0.33 | 0.97 | 3.91 | 2.90 | 0.97 | [63] |
|               |                      | As<sup>3+</sup> | 7.5 | 2.2 | - | - | - | - | - | - | [66] |
|               |                      | As<sup>4+</sup> | 2.5 | 2.3 | - | - | - | - | - | - | [66] |
|               | Poly(itaconic acid/methacrylic acid)-grafted CNC/nanobentonite composite | Ag<sup>+</sup> | 3.5–4.5 | 106 | - | - | - | - | - | - | [61] |
|               |                      | Cd<sup>2+</sup> | 6 | 152 | 259.7 | 2.29 | 0.998 | - | - | - | [62] |
|               |                      | Pb<sup>2+</sup> | 5.5 | 300 | 367.6 | 1.81 | 0.998 | - | - | - | [62] |
|               |                      | Cr<sup>3+</sup> | 6.5 | 2.4 | 2.88 | 1.49 | 0.987 | 0.83 | 0.17 | 0.98 | [65] |
|               | Sodium salt succination (-COO− Na<sup>+</sup>) | Cd<sup>2+</sup> | 6 | 167 | 344.8 | 41.88 | 1.000 | - | - | - | [62] |
|               |                      | Pb<sup>2+</sup> | 5.5 | 300 | 465.1 | 4.13 | 1.000 | - | - | - | [62] |
|               | Poly(itaconic acid/methacrylic acid)-grafted CNC/nanobentonite composite | Co<sup>2+</sup> | 6 | - | 241.8 | 0.025 | 0.99 | 16.31 | 5.04 | 0.98 | [67] |
|               | Xanthate (-ROCS<sub>2</sub>−Na<sup>+</sup>) | Cd<sup>2+</sup> | 6.0 | 24.3 | 154.3 | 0.11 | 0.99 | 26.9 | 2.38 | 0.98 | [68] |
|               | TEMPO-oxidation (-COO−) | Cu<sup>2+</sup> | 6 | 14.6 | - | - | - | - | - | - | [64] |
|               | Periodate/chlorite oxidation | Cu<sup>2+</sup> | 4 | 185 | - | - | - | - | - | - | [69] |
|               | Itaconic acid-grafted-magnetite modified with 2-mercaptobenzamide | Hg<sup>2+</sup> | 8 | 69.7 | - | - | - | 44.05 | 0.36 | 0.991 | [70] |
|               | Carboxylation (-COO−) | Pb<sup>2+</sup> | 7 | - | 2270 | - | 0.978 | - | - | - | [71] |
|               | Amination (NH<sub>3</sub>+) | Cr<sup>6+</sup> | 2.5 | 9.8 | 2.77 | 0.48 | 0.968 | 0.58 | 0.08 | 0.96 | [65] |
|               |                      | As<sup>3+</sup> | 7.5 | 9.3 | 10.56 | 1.85 | 0.991 | 5.52 | 0.22 | 0.986 | [66] |
|               |                      | As<sup>4+</sup> | 2.5 | 9.8 | 12.06 | 5.34 | 0.992 | 6.13 | 0.75 | 0.941 | [66] |
|               | Carboxylated CNC/alginate composite | Pb<sup>2+</sup> | 5.2 | 280 | 339.0 | 0.17 | 0.993 | 68.93 | 0.31 | 0.607 | [72] |
Table 3. Cont.

| Nanocellulose          | Surface Modification       | Heavy Metal | pH  | Removal Efficiency (mg/g) | Langmuir | Freundlich | Ref. |
|------------------------|----------------------------|-------------|-----|----------------------------|----------|------------|------|
|                        |                            |             |     |                            | $Q_{max}$ (mg·g$^{-1}$) | $K_L$ (L·mg$^{-1}$) | $R^2$ | $K_F$ | $1/n$ | $R^2$ |      |
| Unmodified             |                            | Ag$^{+}$    | 5.45| 15.45                      | -         | -         | -     | -     | -     | -     | [60] |
|                        |                            | Cu$^{2+}$   | 6.2 | 13                         | -         | -         | -     | -     | -     | -     | [58] |
|                        |                            | Pb$^{2+}$   | 4.5 | 17.9                       | 24.8      | 0.327     | 0.969 | 0.04  | 0.69  | 0.992 | [73] |
|                        |                            | Zn$^{2+}$   | 5   | 3.5                        | -         | -         | -     | -     | -     | -     | [58] |
|                        |                            | Cr$^{3+}$   | 5   | 14.1                       | -         | -         | -     | -     | -     | -     | [58] |
| Phosphorylation (-PO$_3^{2-}$) |                      | Ag$^{+}$    | 3.5–4.5 | 120                  | -         | -         | -     | -     | -     | -     | [61] |
|                        |                            | Cu$^{2+}$   | 3.5–4.5 | 114                  | -         | -         | -     | -     | -     | -     | [61] |
|                        |                            | Fe$^{3+}$   | 3.5–4.5 | 73                    | -         | -         | -     | -     | -     | -     | [61] |
|                        |                            | Cu$^{2+}$   | 6   | 112                       | -         | -         | -     | -     | -     | -     | [74] |
|                        |                            | 5.7         | 114 | -                         | 18.9      | 0.17      | 0.975 | 6.71  | 0.24  | 0.903 | [72] |
|                        |                            | Ni$^{2+}$   | 6   | 8.6                       | -         | -         | -     | -     | -     | -     | [74] |
|                        |                            | 6.2         | 112 | -                         | -         | -         | -     | -     | -     | -     | [74] |
|                        |                            | Cu$^{2+}$   | 5   | -                         | 19.7      | 0.17      | 0.975 | 6.71  | 0.24  | 0.903 | [72] |
|                        |                            | Ni$^{2+}$   | 6   | 49                        | -         | -         | -     | -     | -     | -     | [58] |
|                        |                            | UO$_2^{2+}$ | 6.5 | 167                       | -         | -         | -     | -     | -     | -     | [56] |
|                        |                            | Pb$^{2+}$   | 6   | 9.7                       | -         | -         | -     | -     | -     | -     | [74] |
|                        |                            | Zn$^{2+}$   | 6.2 | 66.0                      | -         | -         | -     | -     | -     | -     | [74] |
|                        |                            | Cr$^{3+}$   | 6   | 8.9                       | -         | -         | -     | -     | -     | -     | [74] |
|                        |                            | 5           | 58  | -                         | -         | -         | -     | -     | -     | -     | [58] |
|                        |                            | Cd$^{2+}$   | 5   | 5                         | 2.06      | 691       | 0.923 | -     | -     | -     | [76] |
|                        |                            | Cu$^{2+}$   | 5   | 5                         | 1.34      | 3.55      | 0.984 | -     | -     | -     | [76] |
|                        |                            | Cu$^{2+}$   | 5   | 4.7                       | 1.90      | 58.89     | 0.849 | -     | -     | -     | [76] |
|                        |                            | Ni$^{2+}$   | 5   | 4.6                       | 0.74      | 12.32     | 0.964 | -     | -     | -     | [76] |
|                        |                            | Zn$^{2+}$   | 5   | 5                         | 1.61      | 59.21     | 0.902 | -     | -     | -     | [76] |
|                        |                            | Cd$^{2+}$   | 5   | 58.1                      | 405.6     | 61.30     | 0.892 | -     | -     | -     | [77] |
|                        |                            | Cu$^{2+}$   | 5   | 50.6                      | 195.6     | 4.072     | 0.770 | -     | -     | -     | [77] |
|                        |                            | Poly (acrylic acid) grafting | Cu$^{2+}$ | 4.5        | 45.8      | 57.5      | 0.124 | 0.972 | 0.104 | 0.720 | 0.988 | [73] |
|                        |                            | Poly (acrylic acid)/sodium humate grafting | Cu$^{2+}$ | 4.5        | 44.7      | 64.6      | 0.175 | 0.970 | 0.11  | 0.66  | 0.991 | [73] |
|                        |                            | TEMPO oxidation/ polyethyleneimine grafting | Cu$^{2+}$ | 5         | -         | 52.3      | 0.17  | 0.985 | 31.0  | 10.4  | 0.919 | [78] |
|                        |                            | TEMPO-oxidation/GO nanocomposite | Cu$^{2+}$ | 5.7       | 63.5      | -         | -     | -     | -     | -     | -     | [79] |
|                        |                            | TEMPO-oxidation/ nanoGO composite | Cu$^{2+}$ | 5.7       | 68.1      | -         | -     | -     | -     | -     | -     | [79] |
|                        |                            | TEMPO-oxidation (-COO$^-$/thiolation (-Si-SH)) | Hg$^{2+}$ | 5–9       | 145       | 729.9     | 0.36  | 0.998 | 112.0 | 0.43  | 0.835 | [78] |
|                        |                            | Pb$^{2+}$   | 5.5 | 133                       | 137.7     | 0.783     | 0.998 | -     | -     | -     | [80] |
|                        |                            | Cr$^{3+}$   | 4   | 76.5                      | 87.5      | 0.308     | 0.997 | -     | -     | -     | [80] |
Table 3. Cont.

| Nanocellulose | Surface Modification                  | Heavy Metal | pH | Removal Efficiency (mg/g) | Langmuir | Freundlich | Ref. |
|---------------|--------------------------------------|-------------|----|--------------------------|----------|------------|------|
|               |                                      |             |    |                          | $Q_{\text{max}}$ (mg·g$^{-1}$) | $K_L$ (L·mg$^{-1}$) | $R^2$ | $K_F$ | $1/n$ | $R^2$ |
| Poly-methacrylic acid-co-maleic acid grafting | Pb$^{2+}$ | 5 | 165 | - | - | - | - | - | - | - | [81] |
|              | Ni$^{2+}$ | 5 | 117 | - | - | - | - | - | - | - | [81] |
|              | Zn$^{2+}$ | 5 | 138 | - | - | - | - | - | - | - | [81] |
|              | Cd$^{2+}$ | 5 | 135 | - | - | - | - | - | - | - | [81] |
| Bisphosphonate (PO(OH)$_2$) | V$^{5+}$ | 3 | 10.3 | 1.13–1.98 | 1.69–2.92 | 0.97–0.996 | 0.65–1.44 | 0.48–0.987 | [82] |
| Sulfonation  | Pb$^{2+}$ | 5 | 123 | - | - | - | - | - | - | - | [81] |
| Periodate/chlorite oxidation/polyamide-amine-epichlorohydrin | Cu$^{2+}$ | - | 82.19 | - | - | - | - | - | - | - | [83] |
|              | Pb$^{2+}$ | - | 76.11 | - | - | - | - | - | - | - | [83] |
|              | Ca$^{2+}$ | - | 94.77 | - | - | - | - | - | - | - | [83] |
| Unmodified   | Cu$^{2+}$ | 4.5 | 9.67 | 11.2 | 0.011 | 0.962 | 5.389 | 0.13 | 0.838 | [84] |
|              | Pb$^{2+}$ | 4.5 | 22.6 | 24.59 | 0.084 | 0.976 | 5.17 | 0.30 | 0.960 | [84] |
| Phosphorylation (-PO$_3^{2-}$) | Co$^{2+}$ | 4.5 | 4.25 | - | - | - | - | - | - | - | [86] |
|              | Cu$^{2+}$ | 4.5 | 4.77 | - | - | - | - | - | - | - | [86] |
|              | Mn$^{2+}$ | 4.5 | 3.92 | - | - | - | - | - | - | - | [86] |
|              | Zn$^{2+}$ | 4.5 | 4.36 | - | - | - | - | - | - | - | [86] |
|              | Fe$^{3+}$ | 4.5 | 4.19 | - | - | - | - | - | - | - | [86] |
|              | Ho$^{3+}$ | 8 | 13.6 | - | - | - | - | - | - | - | [86] |
|              | La$^{3+}$ | 8 | 10.9 | 12.6 | - | - | - | - | - | - | [86] |
|              | Sm$^{3+}$ | 8 | 12.4 | - | - | - | - | - | - | - | [86] |
| Bacterial nanocellulose | Pb$^{2+}$ | 4.5 | 57.2 | 87.41 | 0.003 | 0.993 | 0.995 | 0.62 | 0.982 | [87] |
| Carboxymethylation (-COO$^-$) | Pb$^{2+}$ | 4.5 | 60.4 | 65.53 | 0.058 | 0.961 | 5.70 | 0.50 | 0.898 | [84] |
| Fe$_2$O$_3$/BC hybrid nanocomposite | Pb$^{2+}$ | 7 | 65 | - | - | - | - | - | - | - | [75] |
| Polyethyleneimine grafting | Pb$^{2+}$ | 5.5 | 28.6 | 125.0 | 0.002 | 0.500 | 1.10 | 0.66 | 0.793 | [85] |
| Fe$_2$O$_3$/BC hybrid nanocomposite | Cr$^{3+}$ | 7 | 25 | - | - | - | - | - | - | - | [75] |
|              | Mn$^{2+}$ | 7 | 33 | - | - | - | - | - | - | - | [75] |

Anionic modification (carboxylic groups) believes to be the most prevalent method to boost the adsorption capacity towards hazardous metal ions removal. The higher numbers of negative binding sites the enhanced complexation and ion-exchange adsorption toward different hazardous metals ions. For example, the carboxylated CNFs can adsorb Pb$^{2+}$, Cd$^{2+}$, Ni$^{2+}$ and Cr$^{3+}$ from aqueous solutions with an efficiency increase of 3–10% compared with unmodified CNFs [74]. Formation of anionic carboxylate groups with quite high densities (TEPO-oxidized) on CNFs has been specifically used for the removal of radioactive uranyl ions (UO$_{2}^{2+}$). The adsorption values of TEMPO-oxidized CNFs are 2–3 times higher than those of traditional adsorbents (hydrogels and SiO$_2$ NPs) [56]. Copper adsorption increased with pH and degree of TEPO-oxidation, with no noticeable adsorption at pH < 7. Since at low pH all carboxylic groups are protonated, and so cannot trap copper ions, as pH increases the volume of carboxylic acid entities decreases to the benefit of their negatively charged carboxylate counterparts, which can adsorb Cu$^{2+}$ [58].
Sulfate functionalization (−O-SO$_3^-$) of nanocelluloses (CNCs and CNFs) surface can be achieved by an acid hydrolysis reaction. Sulfuric acid is a cheap chemical to produce nanocellulose with sulfate groups. However, due to the highly concentrated sulfuric acid, the use of sulfuric acid leads to hydrolysis of the amorphous region and the formation of the CNCs rather than CNFs [57]. Using sulfonated CNF prepared from wheat straw pulp fine, Pb$^{2+}$ was efficiently adsorbed from model solutions (1.2 mmol·g$^{-1}$), whereas unmodified CNF demonstrated low adsorption capacities [54]. The unmodified CNCs had a greater ability to adsorb Ag$^+$ (34 mg·g$^{-1}$) than the CNFs (15 mg·g$^{-1}$) due to the absence of surface sulfate groups [60]. In the remediation of cationic toxic metals from water bodies, CNCs were extracted from rice straw by using a mixture of HCl/H$_2$SO$_4$ acid [59]. The Cd$^{2+}$, Pb$^{2+}$, and Ni$^{2+}$ ions were removed with adsorption capacity of 9.7, 9.42, and 8.55 mg·g$^{-1}$, respectively, at a pH of 6 [61].

Functionalized bacterial nanocellulose (BNC) was investigated for wastewater treatment applications [84,87]. Based on the fact that, specific adsorption capacity (active adsorption sites) of untreated BNC is quite low due to the absence of surface charges [84,86]. The BNC fiber surfaces must be surface functionalized to enhance the specific adsorption and capture of hazardous metal ions and other pollutants. For instance, amino-functionalized BNC, which is synthesized through reaction with epichlorohydin and diethylenetriamine, shows adsorption capacities of 63 for Cu$^{2+}$ and 87 mg·g$^{-1}$ for Pb$^{2+}$ at pH 4.5. However, the grafted amine group protonation at lower pH decreases significantly adsorption. After filtration, the adsorbed metals can be removed by recovery of the aminated BNC during the washes in ethylenediaminetetraacetic acid [87]. BNC chemically modified with ammonium sulfamate (NH$_4$OSO$_2$NH$_2$) displays a maximum adsorption capacity for Pb$^{2+}$ of 22.7 mg·g$^{-1}$, which is five-fold higher than that of unmodified BNC [85]. Likewise, carboxymethylated BNC shows significantly higher adsorption values for Cu$^{2+}$ and Pb$^{2+}$ compared with unmodified BNC (12.6 mg·g$^{-1}$ for Cu$^{2+}$ and 60.4 mg·g$^{-1}$ for Pb$^{2+}$ versus 9.7 mg·g$^{-1}$ for Cu$^{2+}$ and 22.6 mg·g$^{-1}$ for Pb$^{2+}$ at pH = 4.5). These experiments also demonstrated that Cu$^{2+}$ and Pb$^{2+}$ absorption is influenced by variations in pH, contact time, and concentration of the metal ions [84].

Surface grafting of nanocelluloses (CNCs, CNFs, BNC) with functional organic molecules (3-aminopropyl triethoxysilane [77], oxolane-2,5-dione, succinic anhydride [88], ethylenediaminetetraacetic dianhydride [89]) have been recently used to enhance the removal of hazardous metal ions (Ni$^{2+}$, Cu$^{2+}$, and Cd$^{2+}$) from contaminated water. Similarly, grafting of nanocelluloses with natural (chitosan [90], chitin [91], starch [92]) and synthetic (polyethyleneimine [93] and polyacrylamide-acid-co-maleic acid [81]) polymers creates a higher number of active sites with a specific binding affinity for metal ions and consequently enhances adsorption yields [93]. CNF grafting with polyacrylamide-acid-co-maleic acid$^{69}$ and formation of aerogel via radical polymerization improves the adsorption exceeding 95% toward of Pb$^{2+}$, Cd$^{2+}$, Zn$^{2+}$, and Ni$^{2+}$ from wastewater, when their concentration was lower than 10 ppm. Surface grafting CNFs from bamboo with polyacrylic acid increases Cu$^{2+}$ removal from aqueous solutions by 3-fold compared with unmodified CNFs [73]. Recent studies also evaluated the use of synthetic polymers (polyurethane foams, PU) as a template for the dispersion of carboxymethylated CNFs (Figure 3). The composite foams exhibited well dispersed carboxymethylated CNFs in PU matrices with highly porous structure, enhanced mechanical property, and excellent adsorption capacity for Cu$^{2+}$, Cd$^{2+}$ and Pd$^{2+}$ ions removal as well as recyclability [94].

Combination of nanocellulose with other nanoadsorbents (e.g., carbon tube, graphene oxide, zeolites, metal, and metal oxides) have been examined for the capture and elimination of hazardous metal ions from contaminated water [95,96]. Carboxymethylated CNF/hydroxyapatite hybrids have been tested as adsorbents for hazardous metal ions (e.g., absorption capacity for Pb$^{2+}$ ions of ~625 mg·g$^{-1}$) [97]. Cellulose/montmorillonite composites showed high removal capacity (22.2 mg·g$^{-1}$) for toxic bichromate anions (Cr$^{6+}$). Similarly, electrospun cellulose acetate/organically modified montmorillonite showed high removal capacity (~20 mg·g$^{-1}$) for Cr$^{6+}$ removal [98]. Recently, graphene oxide (GO)/dialdehyde matrix...
cellulose membranes cross-linked with triethylenetetramine $[\text{CH}_2\text{NHCH}_2\text{CH}_2\text{NH}_2]_2$ showed maximum adsorption capacity values of 65.1 and 80.9 mg/g for Cu$^{2+}$ and Pb$^{2+}$ ions, respectively [99]. GO was also mixed with cellulose in ionic liquids and was tested for the specific adsorption of Ce$^{3+}$. The hybrid cellulose/GO films, without an obvious interface between phases, display a maximum capacity of 109.1 mg/g for Ce$^{3+}$ adsorption [100]. However, the GO incorporation with cellulose in ionic liquids for water purification applications has been hampered by difficulties due to the small size of GO particles that are difficult to recover [100,101].

Figure 3. Fabrication of a template structure for carboxymethylated (CM) cellulose nanofibers (CNF) with polyurethane (PU) foam with controlled pore structure, for use as a modular adsorbent of heavy metals (Cd$^{2+}$, Cu$^{2+}$, Pd$^{2+}$) in contaminated water [94], ©Elsevier, 2018.

Fabrication of magnetic nanocellulose based adsorbents with the aim of magnetic separation and reuse is a viable approach; it allows processing large effluent volumes and adsorbent regeneration [102]. Recently, hybrid Fe$_3$O$_4$/BNC nanocomposites have been used for the selective removal by magnetic separation of different hazardous metal ions in complex wastewater mixtures and show high adsorption capacity for Pb$^{2+}$ (65 mg·g$^{-1}$), Mn$^{2+}$ (33 mg·g$^{-1}$), and Cr$^{3+}$ (25 mg·g$^{-1}$) [75]. Similarly, aminated BNC/Fe$_3$O$_4$ NPs exhibit high adsorption rates for As$^{3+}$ ions (90 mg·g$^{-1}$) due to their high affinity for magnetic Fe$_3$O$_4$ NPs and amines [103]. A study on BNC composites in which magnetite Fe$_3$O$_4$ NPs were homogeneously distributed in the BNC matrix found that Cr$^{6+}$ ion removal is strongly influenced by the medium pH, with the highest removal efficiency (5.13 mg·g$^{-1}$) at pH 4 [104]. Spherical BNC/Fe$_3$O$_4$ particles, obtained by encapsulating magnetite Fe$_3$O$_4$ NPs of 15 nm in size into BNC particle, showed high adsorption capacities of 65, 33 and 25 mg/g for Pb$^{2+}$, Mn$^{2+}$, and Cr$^{3+}$, respectively [75]. Overall, these studies indicate that magnetic cellulose nanocomposites display excellent adsorption efficiency, compared with individual nanocelluloses.

4. Adsorbents for Hazardous Organic Pollutants Removal

Hazardous organic pollutants (dyes, pharmaceutical compounds, pesticides, fertilizers, and petrochemicals) can pollute water bodies [105,106]. The application of nanocelluloses-based materials (adsorbent, photocatalysts, and filtration membrane) for treating wastewaters contaminated by hazardous organic pollutants has been largely discussed in the literature (Figures 4 and 5), as summarized in Table 4. Generally, the affinity of native cellulose microfibers towards organic pollutants is 100 to 500 times lower than that of
conventional nanomaterials, such as zeolite or activated carbon, due to the low number of active sites for interaction with the organic pollutants [107]. Alternatively, surface-modified nanocelluloses have been tested as support materials for the adsorption of various organic pollutants [39,107–124]. This is mainly explained by their robust mechanical properties, the high specific surface area that allows creating active interaction sites after functionalization, and the small pore size of their filters/membranes. As the nanocellulose intrinsic hydrophilicity is not suitable for the adsorption of organic molecules, surface modifications, and/or formation of nanocomposite materials (e.g., porous films or aerogels with controllable porosity) are required to improve the adsorption and filtration capacity.

**Figure 4.** Highly efficient and selective removal of anionic dyes from water using a composite membrane of cellulose nanofibril (CNF)/chitosan (CS) prepared by de-hydrothermal treatment [125], © Elsevier, 2021.

**Figure 5.** Cellulose nanofibers (CNF) and carbon nanotubes (CNT) or graphene nanoplate (GnP) hybrid aerogels for the adsorption and removal of cationic and anionic organic dyes [126], ©MDPI, 2020.
Table 4. Overview of the different nanocellulose types and their applications as adsorbents to remove organic pollutants.

| Nanocellulose Type | Surface Modification Type | Dye | pH | Removal Efficiency (mg/g) | Langmuir Parameters | Freundlich Parameters | Ref. |
|-------------------|---------------------------|-----|----|---------------------------|---------------------|-----------------------|------|
|                   |                           |     |    |                           | $Q_{\text{max}}$ (mg·g$^{-1}$) | $K_L$ (L·mg$^{-1}$) | $R^2$ | $K_F$ | $1/n$ | $R^2$ |
| Cellulose nanocrystals |                           |     |    |                           |                     |                       |      |
| Sulfate (-SO$_3^-$) |                           |     |    |                           |                     |                       |      |
| Basic fuchsin      |                           | 6–9 | 261 | -                         | -                   | -                     | -    | -    | -     | -     | [118] |
| Crystal violet     |                           | 6–9 | 178 | 185                       | 1.023               | 0.99                  | 44.7 | 0.26 | 0.68  | -     | [127] |
| Methylene blue     |                           | 9   | -  | 118                       | 0.014               | 0.99                  | 18.56| 0.27 | 0.97  | -     | [118] |
| Malachite green    |                           | 6–9 | 178 | -                         | -                   | -                     | -    | -    | -     | -     | [128] |
| Carboxylation (-COO$^-$) |                           |     |    |                           |                     |                       |      |
| Basic fuchsin      |                           | 6–9 | 318 | -                         | -                   | -                     | -    | -    | -     | -     | [118] |
| Crystal violet     |                           | 6–9 | 223 | 243.9                     | 1.129               | 0.986                 | 76.50| 0.21 | 0.803 | -     | [118] |
| Methylene blue     |                           | 9   | -  | 769                       | 0.009               | 0.99                  | -    | -    | -     | -     | [128] |
| Malachite green    |                           | 6–9 | 79  | 101.2                     | 0.019               | 0.974                 | -    | -    | -     | -     | [129] |
| Amination (NH$_3^+$) |                           |     |    |                           |                     |                       |      |
| Acid red GR        |                           | 3.5 | 199.9 | 873.8                  | 0.0305              | 0.92                  | 120.3| 0.34 | 0.985 | -     | [130] |
| Congo red          |                           | 4.7 | 199.5 | -                      | -                   | -                     | -    | -    | -     | -     | [131] |
| Light yellow K-4G  |                           | 4.7 | 183.0 | -                      | -                   | -                     | -    | -    | -     | -     | [131] |
| PVAm grafting      |                           |     |    |                           |                     |                       |      |
| Acid red GR        |                           | 3.5 | 199.9 | 873.8                  | 0.0305              | 0.92                  | 120.3| 0.34 | 0.985 | -     | [130] |
| Light yellow K-4G  |                           | 3.5 | 196.9 | 1211                  | 0.046               | 0.80                  | 237.1| 0.26 | 0.974 | -     | [130] |
| Congo red          |                           | 3.5 | 199.8 | 1619.9                | 0.210               | 0.96                  | 267.3| 0.56 | 0.770 | -     | [130] |
| CNC-infused polyacrylonitrile membrane | |     |    |                           |                     |                       |      |
| Crystal violet     |                           | 7   | 4   | 68                       | -                   | 0.993                 | -    | -    | -     | -     | [132] |
| Keratin composite  |                           | 80  | 485 | 1111                   | 0.043               | 0.995                 | 186.8| 0.28 | 0.944 | -     | [133] |
| Direct Red 80      |                           | 2   | 485 | 1111                   | 0.043               | 0.995                 | 186.8| 0.28 | 0.944 | -     | [133] |
| Reactive Black 5   |                           | 2   | 475 | 1250                   | 0.028               | 0.992                 | 156.8| 0.33 | 0.976 | -     | [133] |
| Alginate hydrogel  |                           |     |    |                           |                     |                       |      |
| Methylene blue     |                           | 7   | -   | 256.4                   | 0.002               | 0.998                 | 1.931| 0.649| 0.988 | -     | [134] |
| Polycracylamide composite | |     |    |                           |                     |                       |      |
| Methylene blue     |                           | 6.5 | 19  | -                       | -                   | -                     | -    | -    | -     | -     | [135] |
| Chitosan-g-poly(acrylic acid) composite (5 wt% CNC) | |     |    |                           |                     |                       |      |
| Methylene blue     |                           | 6   | 1968 | 2074                  | 0.139               | 0.999                 | -    | -    | -     | -     | [136] |
| Imidazolium grafting |                           |     |    |                           |                     |                       |      |
| Orange II          |                           | -   | 98  | -                       | -                   | -                     | -    | -    | -     | -     | [137] |
| Unmodified Quaternization | |     |    |                           |                     |                       |      |
| Methylene blue     |                           | 9   | 100 | 122.2                   | 0.178               | 0.996                 | 12.8 | 0.22 | 0.85  | -     | [138] |
| Crystal violet     |                           | -   | 664 | -                       | -                   | -                     | -    | -    | -     | -     | [139] |
Organic pollutants such as dyes, drugs, and pesticides are toxic even at small concentrations (1 mg L\(^{-1}\)) present in the environment. Organic pollutants in polluted water can cause several health problems to humans including skin rashes, headaches, diarrhoea, muscle and joint pain, difficulty breathing, irregular heartbeat [140,141]. Oral ingestion and inhalation of organic pollutants come under acute toxicity and may lead to different types of cancers, skin irritation, and allergic reactions [142]. Recently, nanocelluloses have been also investigated used as a potential adsorbent to eliminate pesticides, fertilizers, and drugs as contaminants. Bouli et al. (2018) reported that carboxymethyl nanocellulose hydrogels, filled with nanoclays are beneficial adsorbents, soil conditioning agents, and nutrient carriers, especially to improve the growth of cucumber [109]. Likewise, Mautner et al. (2017) demonstrated that the cationic cellulose nanopaper membranes are efficiently useful for the continuous removal of nitrates, released from the chemical fertilizer in the agriculture soil and irrigation water [108]. Similarly, several researches revealed that the nanocellulose can serve as a potential adsorbent for the removal of pesticides, such as chlorpyrifos [143], tetraconazole [144], diuron [145], organophosphorus [146], difenoconazole, and nitenpyram [147]. Nanocelluloses are also employed for the adsorption and elimination of hazardous drugs from industrial wastewater, such as ciprofloxacin, diclofenac [124], metronidazole [148], and ceftriaxone [149]. Nanocelluloses on their own and after anionic functionalization, can be used for cationic organic pollutants adsorption from wastewater. CNFs prepared from kenaf fibers were tested for the adsorption of cationic dyes. The very high adsorption capacity (123 mg/g at pH 9) can be explained by the specific CNF morphology that depends on the selected source [138].

Dyes are particularly dangerous, primarily due to the fast growth of textile manufacturing that consumes about 7 \(\times\) \(10^5\) tons of dyes per year [129]. Dyes are generally non-biodegradable, leading to a severe problem for plants and foods. Direct dyes are water-soluble anionic dyes, have a strong affinity with nanocelluloses. Acid dyes are water-soluble anionic dyes with high very poor affinity to bind nanocelluloses. The attraction forces between the chromophores of direct and acid dyes with nanocelluloses include hydrogen bonding, dipolar forces, and non-specific hydrophobic interactions. Typically, cationic modification of nanocelluloses increases the uptake of the direct and acid dyes. In contrast direct and acid dyes, sulfur dyes (–S–, –S–S–, –Sn–) and azoic (–N=N–), vat dyes are water-insoluble and they are mainly having a high affinity to be adsorbed with nanocelluloses.

Surface modification of nanocelluloses develops functional nanocellulose-based materials and enhances the organic adsorptive property [124,150–152]. To make nanocellulose adsorbents for anionic dye, chemical modifications, such as the introduction of positively charged (cationic modification with amino groups), are usually required (Figure 4). Anionic dyes can be separated using cationic CNFs, obtained by surface quaternization with glycidyl trimethyl ammonium chloride [139]. Several studies focused on the selective adsorption onto cationic CNFs of negatively charged water organic pollutants (e.g., phosphates, nitrates, fluorides, and sulfates) [153]. For instance, CNFs have been grafted with soy proteins to enhance their bioactivity towards biomimetic mineralization. After precipitation of rod-like hydroxyapatite particles, the composite material exhibited high ad-
sorption of methylene blue (up to 454 mg/g), cost-effectivity, and reusability. This suggests that the soy protein-grafted CNF/hydroxyapatite nanocomposite is a promising adsorbent for water purification [119]. A multifunctional Fe-aminoclay/CMC/polyhedral oligomeric silsesquioxane composite was developed to eliminate cationic dyes from contaminated sites. The composite exhibits adsorption of 438 mg/g for methylene blue and 791 mg/g for Chrysoidine G. Its good adsorption capacities are explained by its layered structure and by the presence of amino groups (on the clay surface) and carboxylate/hydroxyl groups (on the CMC backbone) that contribute to the dye adsorption through electrostatic attraction and ion exchange [154].

Combination of the nanocelluloses with inorganic nanoadsorbents (nanoclay, zeolite, nano-hydroxyapatite) has the advantage of eliminating organic pollutants because of the availability of materials as adsorbents [97,121,155]. For instance, Nanohybrids of carboxymethylcellulose (CMC)/hydroxyapatite (HAp) NPs were formed via an in-situ synthetic approach and displayed an adsorption capacity of 200 mg·g⁻¹ for acid yellow 220 removals from aqueous solutions [121]. In another study to increase the functionality of polysaccharides, carboxymethyl cellulose (CMC) grafted with polymethacrylic acid (MAA) was used as a growth modifier to enhance hydroxyapatite mineralization. The CMC/MAA/HAp hybrid material efficiently removes methylene blue with an adsorption value of 671 mg/g [97].

Magnetic nanoparticles can be deposited onto nanocelluloses (CNFs, CNCs, and BC) via in situ hydrolyses of metal precursors that have been investigated for dye removal [156]. The effects of different concentrations of nanocelluloses on the morphology and the magnetic properties of the nanocellulose based materials produced have been discussed in the literature [120,156,157]. For example, spherical magnetic CMC/Fe₃O₄ NPs (NP diameter of 25 nm and CMC from mesquite tree pulp) were investigated as biodegradable adsorbents for organic dyes (methylene blue) in wastewater [120]. Likewise, magnetic nanofibers, prepared by electrospinning cellulose acetate nanofibers and co-precipitation of immobilized ZnO and Fe₃O₄ NPs, show an enhanced capacity to absorb phenol from aqueous solutions (64% in 2 h) [157]. Complex nanocomposites of cyclodextrin-modified CNC in combination with paramagnetic Fe₃O₄/SiO₂ nanorods served as an efficient separation tool for procaine hydrochloride (anesthetic drug) and imipramine hydrochloride (antidepressants drug) [158]. The latter field is believed to become an extremely potential domain that is currently under fast development in view of public health issues.

Highly porous nanocellulose gels (hydrogels, aerogels, organo-gels, sponge-like materials) were prepared from nanocelluloses (CNCs, CNFs, BNC) and their adsorption properties towards a wide range of organic pollutants were investigated [159]. A novel, very robust porous 3D sponge was prepared from reduced graphene oxide (rGO), vitamin C, and CNCs. Vitamin C was used as a reducing agent and as a soft template that allowed creating a unique hierarchical structure with a multitude of interconnected and homogeneously distributed pores, even in the sponge core. This specific structure resulted in a very high surface area for the uptake of methylene blue in water (850 mg/g) [160]. This is much better than what obtained using cellulose/rGO fibers, prepared with the wet-spinning technique (480.8 mg/g) [161]. Hybrid cellulose aerogels assembled from CNFs with carbon nanotubes and graphene nanosheets have been used to remove cationic (methylene blue) and/or anionic (Congo red) dyes [126]. Comparison of the hybrid aerogel structural properties and adsorption mechanics in the function of the CNF/GnP ratio indicated that the optimum performance is obtained with a CNF/graphene nanosheets ratio of 3:1. For the single dye system, the adsorption kinetics for methylene blue (maximum adsorption: 1178 mg·g⁻¹) and Congo red (maximum adsorption: 585 mg·g⁻¹) are explained by pseudo-second-order adsorption kinetic and the monolayer Langmuir adsorption isotherm. For the binary dye system, the CNF/graphene nanosheets hybrid displays better dye adsorption capacity compared with pristine CNFs and graphene nanosheets. Moreover, almost 80% of methylene blue or Congo red can be desorbed from CNF/GnP using ethanol as a desorption agent. In conclusion, aerogels from nanohybrid materials made of CNF with carbon
nanotubes or GnP are promising reusable adsorption materials for dye removal (Figure 5). Table 4 gives an overview of the different nanocellulose types and surface modification procedures, highlighting the variety of adsorption material for organic dyes [126].

5. Wettable Materials for Oil/Water Separation

Oil-based compounds from chronic discharges and oil spills represent an important source of environmental pollution. Oil toxicity is mostly related to the aromatic fraction made of both low molecular weight (LMW)/high molecular weight (HMW) polycyclic aromatic hydrocarbons (PAHs) ratio [162]. As well, dissolved fractions of petroleum appear to be more bioavailable to aquatic organisms. Techniques to separate oil/water mixtures are crucial for many industries (e.g., food, pharmaceutical, and petrochemical) that use and produce huge amounts of oils and require solutions for their removal from wastewater [163]. Normally, efficient water/oil separation is accomplished by separating water and oil, followed or in combination with oil adsorption. Currently, the most common water/oil separation methods (sedimentation, centrifugation, or hydrocyclone) are based on differences in gravity or density [163,164]. Recently, nanocelluloses (CNFs and CNCs) incorporation in membranes for oil separation offers interesting opportunities, together with the (partial) modification of the original hydrophilic surface properties towards higher hydrophobicity and/or balanced amphiphilicity [165]. Besides CNFs and CNCs, electrospun cellulose nanoyarn membranes with grafted hydrophobic moieties have been used for oil removal. The step-by-step fabrication of oil/water separation membranes by electrospinning of cellulose acetate, followed by their deacetylation, is shown in Figure 6 [163]. Interestingly, electrospun cellulose acetate nanofibers are super-amphiphilic in air, oleophobic in water, and super-hydrophilic in oil. The membranes are characterized by high separation flux (up to 38,000 L/m²·h) and gravity-based separation efficiency (up to 99.97%) for chloroform/water mixtures. Moreover, their anti-pollution and self-cleaning capacities boost their recyclability. Therefore, cellulose acetate nanofiber membranes could be interesting for chemical plants, textile mills, food industry, and also offshore oil spills [163].

Highly porous nanocellulose gels and sponge-like materials prepared from nanocelluloses (CNCs, CNFs, BNC) are frequently exploited for oil/water separation due to their high oil adsorption capacity thanks to their porous structure [165]. For example, CNFs can be modified with silanes to make them hydrophobic and to adsorb oil contaminants from the water surface [166]. Likewise, CNF aerogels coated with a thin layer of TiO₂ NPs by sol-gel chemistry are efficient for oil adsorption [167]. Moreover, silylated nanocellulose sponges provide a flexible, hydrophobic, and ultra-light structure for oil removal from water mixtures [164]. These highly porous (up to 99%) silylated CNF sponges were obtained, using a freeze-drying method, from the pulp powder of oat straw cellulose through reaction with methyltrimethoxysilane. These sponges are more flexible than common inorganic porous materials, as indicated by the 96% of maximum shape recovery after compressive deformation. Their oleophilic and hydrophobic features allow the efficient and selective removal by adsorption of dodecane spills from surface water [164]. Similarly, ultralight carbon aerogels prepared from hydrophobized cellulose microfibers can be valuable for selective oil sorption [168]. These sponges show 99% of porosity, 0.01 g/cm³ density, good hydrophobicity, reusability, and very high adsorption capacity (>86 g/g) towards paraffin oil. Surface hydrophobization of BNC membranes has been modified with trimethylchlorosilane for the efficient removal of plant oil from water [169], by dipping BNC aerogels into liquid phase trimethylchlorosilane followed by freeze-drying (Figure 7). The obtained very hydrophobic (water/air contact angle as high as 146.5°) and highly porous (∼99.6%) surface-modified BNCs offer high selectivity for oil adsorption from water, with absorption capacities up to 185 g/g [169].
Figure 6. Formation of dual super-amphiphilic modified cellulose acetate nanofiber membranes by electrospinning, with highly efficient oil/water separation and excellent antifouling properties, (a) Electrospinning process to prepare cellulose acetate nanofiber membrane; (b) Deacetylation process to form d-CA; (c) Prewetted process to form amphiphilic structure; and (d) super-amphiphilic structure to separate water and oil [163]. ©Elsevier, 2020. CA, cellulose acetate nanofiber; d-CA, dual super-amphiphilic modified cellulose acetate nanofiber.

Figure 7. Surface hydrophobization of bacterial cellulose acetate membranes with trimethylchlorosilane for the efficient removal of plant oil from water [169], ©American Chemical Society (2015). BCA—bacterial cellulose acetate; TMCS—trimethylchlorosilane; HBCA—hydrophobic BCA.
Incorporation of magnetic nanoparticles onto nanocelluloses via blending or in situ hydrolysis of metal precursors that have been used for oil removal from wastewater. Magnetic nanocellulose aerobeads, fabricated by freeze-drying of iron oxide (Fe$_3$O$_4$)-containing spherical CNFs (from used cardboard boxes), also show very good selectivity towards oil removal. These aerobeads (0.005 g/cm$^3$ of density and 99% of porosity) possess excellent absorption efficiency towards various oils and organic solvents, especially castor oil (279 g/g). They also show superior recyclability and can be reused at least 10 times with high absorption capacity towards diesel oil (101 g/g) [170]. Fe$_3$O$_4$-based nanocellulose aerogels fabricated using the freeze-drying method can be used for the absorption of spilled oil in water. These aerogels have a density of 9.2 mg/cm$^3$ and 68 g/g of adsorption capacity towards cyclohexane, with good absorption also of ethyl acetate, and vacuum pump oil [171]. This study indicates that magnetic nanocellulose aerogels and their nanocomposites can efficiently adsorb and eliminate oil from contaminated water; however, their biocompatibility and environmental degradability must be thoroughly investigated before their utilization for large-scale and long-term applications.

6. Flocculants and Coagulants for Suspended Materials

Flocculants are agents that can promote flocculation of colloids and suspended particles in liquids to aggregate them for floc formation [172,173]. Nanocelluloses such as CNF and CNC their modified counterparts have been investigated as flocculants for the treatment and elimination of contaminants from wastewater. These nanoflocculants induce flocculation of the suspended particles in contaminated wastewater by neutralizing the surface charge of particles or by forming bridges between the individual suspended particles. Figure 8 explains how anionic nanocelluloses (CNFs and CNCs) function as flocculants for the capture and removal of charged pollutants [10]. In contrast to native cellulose, there are several characteristic features of nanocelluloses (CNFs and CNCs) that make them an ideal flocculants candidate for water treatment: (1) small size and high-surface-area rod-like morphology that give rise to percolation at low concentrations; (2) CNFs and CNCs can improve the formation of flocs compared to native fibers. In comparison between CNFs and CNCs, the higher electrostatic repulsion and rigidity of CNCs than CNFs will prevent the occurrence of physical and chemical entanglements contributing to reduced risk of gelation.

![Figure 8](image-url)

**Figure 8.** Flocculation mechanism of anionic nanocellulose to remove pollutants from water. (A) Binding and flocculation of cationic pollutants, and (B) visual observation of flocculation efficiency [10], ©Springer, 2017.
To date, there are only very few studies describing the applications of nanocelluloses (CNCs and CNFs) as flocculants in wastewater treatment; some examples are given in Table 5. Suopajarvi et al. (2013) fabricated carboxylated CNFs as anionic flocculants for municipal wastewater treatment. The high and long-lasting stability of anionic CNFs in aqueous suspensions provided excellent performance (turbidity reduction of 40–80% and COD removal of 40–60%) within the desirable pH range from 6 to 8 [174]. Likewise, Korhonen and Laine (2014) examined CNC/polyelectrolyte with different charge density for retention and flocculation of kaolin and calcium carbonate fillers in the papermaking industry. They showed that the flocculation efficiency is increased from 80% to 95% in the case of CNF/polyelectrolyte. The presence of polyelectrolytes induces the formation of CNF/polyelectrolyte bridges between kaolin and calcium carbonate particles and leading to efficient flocculation [175]. A recent study assessed the flocculation performance of hyperbranched cellulose grafted with polyethyleneimine \((C_2H_5N)_n\) for the treatment of kaolin-contaminated wastewater. This cellulose-based flocculent decreased the residual turbidity of kaolin suspension from original 490 NTU to 4 NTU under 2.4 mg/L of the flocculent at pH 7.0 for 30 min [176].

Recently, Kemppainen et al. (2016) produced sulfonic acid and dicarboxylic acid cellulose (anionic) CNCs for the flocculation of quartz and hematite suspensions in contaminated water. The most effective performance was obtained at a pH of 8–9, and a carboxylic modified CNFs at dosage of 200–500 ppm was enough to flocculate hematite efficiently. The sulfonated modified CNFs is efficient a hematite flocculant as carboxylic modified CNFs at a dosage of 500 ppm after longer conditioning time and less vigorous stirring [177]. Campano et al. (2019) fabricated cationic CNCs as a novel flocculant for kaolinite/clay suspensions. The fastest flocculation values (10–30 mg/g) and biggest floc size were near the isoelectric point [178]. Yu et al. (2016) used microcrystalline cellulose (MCC) to fabricate carboxylated CNCs (length of 200–250 nm and diameter of 15–20 nm) by citric-hydrochloric acid hydrolysis. They could use these CNCs as a flocculant to remove cationic dyes and kaolin from suspensions with 99.5% of turbidity removal capacity [128].

### Table 5. Various nanocelluloses-based flocculants used for the water treatment process.

| No. | Nanocellulose Flocculants | Contaminants | Optimum Flocculation Conditions | Highest Removal Efficiency | Ref. No. |
|-----|---------------------------|--------------|---------------------------------|---------------------------|----------|
| 1   | Pristine CNCs             | *Pseudomonas aeruginosa* (Gram-negative Bacteria) | Flocculant concentration (CNC:Bacteria) = 100,000:1, reaction time = 24 h | 100% | [179] |
| 2   | Carboxylated CNCs         | Kaolin clay (suspended filler particles) | Flocculant concentration = 40 mg L\(^{-1}\), pH = 4–10, reaction time = 30 min | 95.4% | [128] |
| 3   | Pyridinium grafted CNCs    | *Chlorella vulgaris* (Microalgae) | Flocculant concentration = 0.1 g flocculant per g microalgae, pH = 4–11, reaction time = 30 min | 95% | [180] |
| 4   | Amine-functionalized CNCs  | Sodium dodecyl sulfate (anionic surfactant) | Optimum pH = 4, | - | [181] |
| 5   | Carboxylated CNFs         | Suspended particles | Flocculant concentration = 2.5–5.0 mg dm\(^{-3}\), pH = 6–8, reaction time = 30 min | 40–80% | [174] |
| 6   | Sulfonated CNFs           | Suspended particles | Flocculant concentration = 2.5 mg dm\(^{-3}\), reaction time = 30 min | - | [182] |
| 7   | Quaternized CNFs          | Reactive orange 16 (reactive dye) | Flocculant concentration = 150 mg·L\(^{-1}\), reaction time = 12 h \(q_{max} = 0.477 \text{ mmol·g}^{-1}\) for the reactive dye | | [183] |
7. Photocatalytic Materials for Hazardous Pollutants Degradation

Photocatalysis has emerged as an environmentally friendly, low-cost, and handy approach for the degradation of organic pollutants and/or dyes in wastewater. Even though nanocelluloses alone exhibit limited photocatalytic activity under the visible light-UV region, conventional metal oxide photocatalyst (ZnO [184] and TiO$_2$ [185]) have been added to enhance the photocatalytic activity. These photocatalytic materials (individual particles, thin-film, membranes) have been developed using cellulose-based metal oxide nanostructures in the form of under UV and visible light irradiation. Photocatalysis harnesses photon energy to produce active radicals that promote the decomposition of organic pollutants [186]. When the photon energy is above the semiconductor energy gap, semiconductor materials can generate active hydroxyl radicals, leading to the excitation of electrons from the valence band into the conduction region [187]. This creates a hole that can generate hydroxyl radicals in an alkaline medium [188]. These radicals are highly reactive and can enhance the oxidation of recalcitrant chemicals in wastewater, leading to complete degradation of organic pollutants into nontoxic byproducts (i.e., CO$_2$ and H$_2$O) [189]. Active hydroxyl free radical radicals attack organic materials via four known pathways: radical addition, hydrogen abstraction, electron transfer, and radical combination (Figure 9a) [190]. The main challenge associated with semiconductor NP use in photocatalysis is their removal from the liquid after the reaction. Poor visible light absorption nanocellulose/TiO$_2$ and nanocellulose/ZnO is the limitation of such photocatalysts due to the wide bandgap. Similarly, separation of these nanocomposites is difficult from reaction mixture due to non-magnetic characteristics.

![Figure 9. (a) Mechanism for photocatalytic treatment and degradation of an organic pollutant (methylene blue) in wastewater using graphene oxide (GO)/cellulose/TiO$_2$ nanocomposites [190], ©Royal Society of Chemistry, 2020. (b) Deposition of TiO$_2$ nanotubes and AgO$_x$ nanoparticles (NPs) onto cellulose nanofibers (CNFs) for improved photocatalytic cleaning of wastewater [191], ©Springer, 2019.](image-url)
Narrow bandgap (2–2.4 eV) semiconductors such as CuO$_2$, CdS, Fe$_2$O$_3$ are recommended to generate more electron–hole pairs under visible light [192]. Incorporating co-catalyst NPs (Ag, AgOx, graphene) can narrow the bandgap and further improve the photocatalytic performance of wide bandgap photocatalysts in the visible region (Figure 9) [193,194]. Ferromagnetic photocatalysts such as Fe$_3$O$_4$ NPs can give the required optical absorption at the visible region with relatively narrow band gaps (~2.1 eV). More interestingly, their nanocomposite with nanocelluloses can be easily recovered from wastewater for recycling and further use, thanks to their unique magnetic properties [195]. Jiao et al. (2018) showed that a magnetic Fe$_3$O$_4$/CNF composite aerogel is an effective catalyst for the Fenton-like degradation of Rhodamine B (100% of removal efficiency) [196]. This catalyst was re-used for six successive runs and retained 97% of its removal capacity [197]. Novel cellulose-based photocatalysts have been developed with appropriate band gap (2.4 eV) and excellent photocatalytic activity and stability under UV-Vis light [198]. BiOBr exhibit narrow bandgap of 2.4 eV and high phenol degradation rate under visible light when using BiOBr/regenerated cellulose than BiOBr particles (80% versus 45% after 3 h of irradiation) [198].

Porous nanocellulose materials (cellulose aerogels and cellulose sponge) loaded with photocatalyst NPs have been also transformed into photocatalytic carbonaceous materials by hydrothermal treatment followed by thermal pyrolysis, among others [199]. The photocatalytic properties of such photocatalytic carbonaceous materials are enhanced because of their high surface area and porosity as well as their high ability for adsorbing organic pollutants [200]. Photocatalytic cellulose-based nanopapers have been also investigated as a novel support material for photocatalyst NPs immobilization. Matsubara et al. (1995) prepared TiO$_2$-containing papers for the catalytic degradation of acetaldehyde under ultraviolet light [178]. Electrosprun cellulose acetate membrane loaded with TiO$_2$ (5 wt.%) show very high dye removal by photocatalytic degradation [185]. The resulting TiO$_2$/cellulose composites efficiently degrade phenol (90% after 4 h) under weak ultraviolet light irradiation [195]. Table 6 lists the various nanocellulose/inorganic composites that have been used for wastewater treatment by adsorption or photocatalytic degradation.

| Cellulosic Material | Inorganic | Preparation Method | Removal Process | Pollutants | Removal Capacity | References |
|---------------------|-----------|--------------------|-----------------|------------|-----------------|------------|
| Cellulose-graft-soy protein isolate | Calcium phosphate–rod-like shape hydroxyapatite | Bio mineralization | Adsorption | Methylene blue | 454 mg/g | [161] |
| Carboxymethyl cellulose | Hydroxyapatite | In situ precipitation | Adsorption | Pb$^{2+}$ acid yellow 220 | 625 mg/g and 200 mg/g | [97] |
| Carboxymethyl cellulose-graft-poly(methacrylic acid) | Rod-like hydroxyapatite | Bio mineralization | Adsorption | Methylene blue | 671 mg/g | [121] |
| Carboxymethyl cellulose | Carboxymethyl cellulose, hydroxyapatite and lysine membrane | Conventional casting or double decomposition methods | Adsorption | Bisphenol A | - | [201] |
| Cellulose | Montmorillonite | - | Adsorption | Cr (VI) | 22.2 mg/g | [21] |
| Cellulose acetate | Organically modified montmorillonite | - | Adsorption | Cr (VI) | - | [98] |
| Cellulose and carboxymethyl cellulose | Clay | Crosslinking in NaOH/urea aqueous solution | Adsorption | Methylene blue | 98% | [202] |
| Cellulose | GO/cellulose membranes | Pressed membranes | Adsorption | Heavy metals | - | [203] |

Table 6. Nanocellulose/inorganic composites as effective wastewater cleaning agents.
| Cellulosic Material          | Inorganic | Preparation Method                             | Removal Process         | Pollutants                | Removal Capacity | References |
|-----------------------------|-----------|-----------------------------------------------|-------------------------|--------------------------|------------------|------------|
| Dialdehyde cellulose        | GO        | Crosslinked with triethylene-tetramine       | Adsorption              | Cu$^{2+}$ and Pb$^{2+}$ | 65.1 and 80.9 mg/g | [102]      |
| MCC                         | GO        | GO/microcrystalline cellulose aerogels prepared using lithium bromide | Adsorption              | Methylene blue           | 2630 mg/g        | [204]      |
| CNC                         | rGO       | 3D sponge using vitamin C                     | Adsorption              | Methylene blue           | 850 mg/g         | [160]      |
| Cellulose                   | GO        | Wet-spinning technique                        | Adsorption              | Methylene blue           | 481 mg/g         | [161]      |
| Bacterial cellulose         | GO        | Composite membrane                            | Selective ion permeation| Different inorganic/organic ions | -                | [205]      |
| Cellulose                   | GO        | Using ionic liquids as solvent                | Adsorption              | Ce$^{3+}$                | 109.1 mg/g       | [205]      |
| Cellulose                   | CNTs      | Grafting Welan gum onto CNTs entrapped cellulose beads | Adsorption              | Methylene blue           | 300 mg/g         | [206]      |
| CNCs                        | Carbon monolith | -                           | Adsorption              | Methylene blue           | 127 mg/g         | [129]      |
| Bacterial cellulose         | Fe$_3$O$_4$| Cellulose-magnette composites                | Adsorption              | Cr(VI)                   | -                | [104]      |
| Bacterial cellulose         | Fe$_3$O$_4$| Agitation fermentation                       | Adsorption              | Pb$^{2+}$, Mn$^{2+}$ and Cr$^{3+}$ | 65, 33 and 25 mg/g | [75]      |
| Mesquite tree pulp          | Fe$_3$O$_4$| Spherical magnetic NPs through co-precipitation | Adsorption              | Methylene blue           | 1225 mg/g        | [120]      |
| Cellulose acetate fibers    | Zno NPs with Fe$_3$O$_4$| Coprecipitation                  | Adsorption              | Phenol                   | 64%              | [157]      |
| Carbon microspheres         | Magnetic CoFe$_2$O$_4$| -                             | Photodegradation         | Rhodamine B              | -                | [194]      |
| Cellulose                   | Fe$_3$O$_4$| Co-precipitation                             | Photodegradation         | Methylene blue           | -                | [192]      |
| Cellulose                   | Fe$_3$O$_4$| Hydrothermal technique                       | Photodegradation         | Rhodamine B              | 100%             | [196]      |
| Carboxyl cellulose          | Fe$_3$O$_4$| Chemical co-precipitation technique           | Photodegradation         | Navy blue                | -                | [207]      |
| Amidoximated bacterial cellulose | ZnO   | In situ polyol method                         | Photocatalytic property  | Methyl orange            | 92%              | [184]      |
| Cellulosic fabric           | ZnO       | Sol-gel process                               | Photodegradation         | Methylene blue           | -                | [208]      |
| Cellulose                   | ZnO       | Co-precipitation                              | Photodegradation         | Methylene blue           | 69.5%            | [209]      |
| Cellulose                   | TiO$_2$   | Acids                                        | Photodegradation         | -                        | -                | [210]      |
| Hydroxpropyl methyl cellulose | TiO$_2$| Nano-photocatalysts prepared in situ          | Photodegradation         | 4-nitrophenol            | -                | [211]      |
| Bacterial cellulose         | TiO$_2$   | -                                            | -                        | Pb                       | 90%              | [212]      |
| CMC                         | TiO$_2$   | Polyamine/CMC/TiO$_2$ nanocomposite           | Adsorption               | Congo red                | 94.3 mg/g        | [213]      |
| Paper                       | TiO$_2$   | -                                            | Catalytic degradation    | Acetaldehyde             | -                | [214]      |
8. Membrane Materials for Wastewater Treatment

Membrane technology has emerged as a favorite processing and separation method (filtration, extraction, and distillation) for reclaiming water from salty and contaminated water streams for their re-use by humans. The use of nanocellulose based membranes for water purification and/or desalination is a highly efficient and environmentally responsible method because of the low energy consumption and high selectivity. The operational characteristics of nanocellulose based membranes are mostly determined by two parameters: their water permeation flux value, and their specific pollutant removal capacity. The nanocellulose pore size and surface conditions are important parameters for controlling the filter efficiency. The membrane rejection might be explained by two mechanisms: (i) size-exclusion effects, and (ii) the membrane chemical or physical affinity for the pollutant. In the first mechanism (size-exclusion), the pollutant is selectively rejected depending on its size relative to the membrane pore size. The functioning and efficiency of nanocellulose based membranes depend on their selective retention of pollutants. Depending on the pollutant size, cellulose-based membranes are categorized into [195]:

1. Microfiltration membranes with microspores of 10 μm to 100 nm; to be used for removal of suspended particles and Bacteria (200 nm–30 micron)
2. Ultrafiltration membranes with nanopore range from 100 nm to 2 nm; it is suitable for removal of nanoparticles and viruses with sizes 50–200 nm
3. Nanofiltration membranes with nanopore size range from 2 nm to 1 nm; to be used for removal of organic pollutants (0.7–1.5 nm)
4. Reverse osmosis with sub-nanopore size from 0.1 to 1 nm; to be used for hazardous metal ion (0.1–0.7) removal.

Nanocellulose membranes predominantly operate according to the size-exclusion mechanism, and the pore size depends on the formation of a random CNF or CNC network that generates the “micro/nano-pores”. The pore size (micro/nanosize) can be modulated through variations in the aspect ratio and choice of nanocellulose material (CNFs/CNCs) and processing steps (thickness and dense packing). However, it is worth noting that small pore sizes generate higher pressures during the transportation of the effluent flow through the membrane [11]. Based on a theoretical pore flow model, pollutants with sizes larger than the mesh of the nanocellulose based membranes are captured by the size exclusion mechanism. Conversely, the rejection mechanism for smaller pollutants is mainly governed by the pollutant solution-diffusion mode. This does not often occur with nanocellulose based membranes. The second removal mechanism (affinity) depends on the electrostatic interactions between the pollutant and the surface functionalities of the nanocellulose based membranes. The surface modifications have been successfully used to tune nanocellulose reactivity towards different water pollutant types (e.g., biological species, inorganic pollutants, heavy metals). The modification processes include and are not limited to: (1) carboxylic acid halides create ester linkages, (2) sulfuric acid treatment provides sulfate esters, (3) epoxides create ether linkages, (4) acid anhydrides create ester linkages, (5) TEMPO mediated hypochlorite oxidation creates carboxylic acids, (6) isocyanates create urethane linkages, (7) chlorosilanes create an oligomeric silylated layer; (8) halogenated acetic acids create carboxymethyl surfaces [216]. The chemical surface modifications modulate the nanocellulose hydrophobic and hydrophilic properties because its intrinsic
Hydrophilicity contributes to the swelling and loss of mechanical strength or stiffness of the filter material. High hydrophobicity might increase the flow resistance and/or modify the selectivity for the adsorbed species.

To date, three distinct techniques have been investigated for the fabrication of nanocellulose membranes: (i) electrospinning of cellulose mats and impregnation [217], (ii) vacuum filtration of the nanocellulose suspension and coating [218], and (iii) freeze-drying of the CNF suspension in the presence of an additional matrix component [131]. In parallel with papermaking, vacuum filtration is the most straightforward and feasible method for scaling up and for the fabrication of nanopaper membrane that can then be deposited onto a carrier substrate or used as free-standing units [218]. The final structure can be stabilized, in combination with hot-pressing to add a matrix material, to fabricate fully bio-based membranes. The obtained nanopapers can be used as membranes to efficiently adsorb organic compounds, phosphates, nitrates, fluorides, metal ions, or biological substances (e.g., bovine serum albumin) [219]. The efficiency of nanocellulose membrane can be enhanced by adding specific chemical modifications (e.g., coating, grafting), and/or by deposition of functional NPs. Figure 10 shows enhanced performance of nanocellulose membranes for forwarding osmosis through modification of the nanocellulose membrane by coating with a polymer (e.g., polyamide active layer), and by nanoparticle (NP) deposition (e.g., Ag, Pt) [220].

Electrospun cellulose nanofiber membranes (ENMs) are a cutting-edge membrane technology that offers a high rejection rate compared to conventional membranes. As shown in Figure 11, hollow Fe(OH)$_3$/cellulose can be obtained by electrospinning using a suitable solvent system, whereby the solvent exchange mechanism is induced and solid or free core structures are created [221]. Concomitantly, Fe(OH)$_3$ NPs are formed and deposited onto the cellulose membrane structure during the in situ reductions of FeCl$_3$ added to the spinning solution. The fiber size and diameter can be controlled by modifying the spinning parameters (e.g., applied voltage and concentration ratios). Moreover, increasing NaOH concentration in the coagulation bath leads to larger fiber diameters and surface pore sizes. The hybrid nanocellulose fibers fabricated with this method display extremely
specific surface area and many active species for the interaction with effluents, and consequently good removal activity of organic dyes [221]. Similarly, Yang et al. (2014) developed composite membranes from CNFs functionalized with thiols that were dimensionally stabilized and fixed after cross-linking into an electrospun polycrylonitrile nanofibrous scaffold during thermal curing. They tested these membranes in an ultrafiltration set-up for $\text{Cr}^{6+}$ and $\text{Pb}^{2+}$ removal with satisfactory results [222]. Similarly, Wang et al. (2013) prepared nanofibrous membranes containing CNCs by electrospinning to remove Brevundimonas diminuta and Escherichia coli bacteria from water in conditions of relatively high flux rate and low drop in pressure [223].

![Figure 11](image.png)

**Figure 11.** Electrospinning with suitable solvent systems was used to fabricate hybrid nanocellulose fibers with solid or hollow core and deposition of $\text{Fe(OH)}_3$/cellulose nanoparticles: (a) Schematic representation of the electrospinning process for nanofiber fabrication; (b,c) Schematic representation of the hollow fibers as membranes for diffusion and regeneration, respectively; (d-f) Photographs of the $\text{Fe(OH)}_3$@Cellulose HNFs membranes and the dead-end filtration device, [223] ©American Chemical Society, 2017.

Limiting fouling by the addition of biological and/or organic moieties is a crucial issue to ensure the nanocellulose membrane’s durability. CNFs and CNCs hydrophilicity and easily functionalizable surface are important features to retard and/or prevent fouling [224]. Membranes prepared by CNF incorporation into composites with a polymer matrix, such as cellulose acetate [225,226], polyethylene oxide [227], polyvinyl alcohol [228,229] polypyrrole [230], poly(ether sulfone), and polyhydroxybutyrate [231], have been tested for the removal of heavy metals. Moreover, composite membranes obtained by incorporating zeolitic imidazolate framework-8 (ZIF-8) in CNFs show high durability for the selective
removal of 99% of cationic dyes from anionic and cationic dye mixtures (Figure 12). Likewise, CNF filter papers from microfibrillar Cladophora cellulose have been tested for the removal of viruses (e.g., xenotropic murine and swine influenza viruses) and also of Au NPs from aqueous solutions [232–234].

![Figure 12. Control of the cellulose nanofibrous composite membrane structure by in-situ synthesis of zeolitic imidazolate framework-8 (ZIF-8) at cellulose nanofibers (CNF) for the selective removal of cationic dyes [235], © Elsevier, 2019.](image)

Bacterial cellulose-based membranes have been developed and their performance was compared with commercial membranes for water pollutants removal. BNC nanopaper sheets have been used as ultrafiltration membranes with more than 75% of rejection efficiency towards polyethylene glycol. BNC rejection efficiency can be further increased to 85 to 90% by lamination with β-chitin or deacetylated chitin sulfonate composites [236]. The combination of BNC membranes and cross-linked CNF offers high separation efficiency (>98%) of stabilized or non-stabilized oil-in-water emulsions, with droplet diameters below 1 µm [7]. Other filtration features for the removal of specific components can be added to nanocomposite BNC membranes. For instance, BNC and GO are combined into a nanocomposite membrane by diffusing the GO into a BNC formamide gel. The freestanding BNC/GO membranes display specific permeation for distinct inorganic/organic ions with variable sizes (from the nano-scale to molecular-scale ranges) [205], enhanced mechanical strength and toughness, and high stability against swelling or water environments. Table 7 provides an overview of the use of nanocelluloses for the fabrication of filtration membranes and their contribution to wastewater treatment.
Table 7. Filtration membranes are made of nanocelluloses for wastewater treatment. CA, cellulose acetate; TMP, transmembrane pressure; PAE, polyamideamine-epichlorohydrin; PAN, polyacrylonitrile; PET, polyethylene terephthalate.

| Nano Celluloses | Membrane                  | Pollutants | Thickness (µm) | Porosity (%) | Flux (L.h⁻¹.m⁻²) | TMP (kPa) | Permeability (L.h⁻¹.m⁻².Pa⁻¹) | Efficiency (% or mg·g⁻¹) | Ref. |
|-----------------|---------------------------|------------|---------------|--------------|------------------|----------|---------------------------------|--------------------------|-----|
| Unmodified CNC  | PEG                       | 20–60      | 35            | -            | -                | 200–500  | 1.8 × 10⁻⁵–6.1 × 10⁻⁵           | 20–75% [237]             |     |
|                 | Oil                       | 0.6–10     | -             | -            | 1036–1734        | 50       | 2.1 × 10⁻²–3.5 × 10⁻²           | 99.9% [238]              |     |
| CNC-coated CNF  | Ag⁺⁺, Cu⁺⁺, Fe²⁺, Fe³⁺   | 201–210    | 22–23         | 0            | 450              | 0        | -                              | 91–94% [239]             |     |
| Acetone treated |                           |            |               |              |                  |          |                                 |                          |     |
| CNC-coated CNF  |                          |            |               |              |                  |          |                                 |                          |     |
| Oxidized CNC    |                           |            |               |              |                  |          |                                 |                          |     |
| CNC/gelatin-coated cellulose sludge/CNF | | | | | | | | |
|                |                          |            |               |              |                  |          |                                 |                          |     |
| Acetone treated |                           |            |               |              |                  |          |                                 |                          |     |
| CNC-coated CNF  |                          |            |               |              |                  |          |                                 |                          |     |
| Oxidized CNC    |                           |            |               |              |                  |          |                                 |                          |     |
| CNC/gelatin-coated cellulose | | | | | | | | |
|                |                          |            |               |              |                  |          |                                 |                          |     |
| Cellulose nanocrystals | | | | | | | | |
| Phosphorylated | Methyl violet 2B          | 440–448    | 65            | -            | 50–150           | 9.5 × 10⁻³ | 0.33 mg·g⁻¹                      | 0.33 mg·g⁻¹ [241] |     |
| CNC/chitosan    |                           |            |               |              |                  |          |                                 |                          |     |
| Cross-linked    |                           |            |               |              |                  |          |                                 |                          |     |
| CNC/chitosan    |                           |            |               |              |                  |          |                                 |                          |     |
| Cross-linked    |                           |            |               |              |                  |          |                                 |                          |     |
| CNC-chitosan    |                           |            |               |              |                  |          |                                 |                          |     |
| CNC-impregnated | Victoria blue 2B         | 52–116     | 69–73         | 17,838       | 600–800         | 2.069 × 10⁻² | 52.6–99.9%                     |                          |     |
| electrospun CA  |                           |            |               |              |                  |          |                                 |                          |     |
| CNC/chitosan    |                           |            |               |              |                  |          |                                 |                          |     |
| Cross-linked    |                           |            |               |              |                  |          |                                 |                          |     |
| CNC/chitosan    |                           |            |               |              |                  |          |                                 |                          |     |
| Cross-linked    |                           |            |               |              |                  |          |                                 |                          |     |
| CNC-impregnated |                           |            |               |              |                  |          |                                 |                          |     |
| electrospun PAN |                           |            |               |              |                  |          |                                 |                          |     |
| Oxidized CNC    |                           |            |               |              |                  |          |                                 |                          |     |
| CNC-impregnated |                           |            |               |              |                  |          |                                 |                          |     |
| Electrospun CA  |                           |            |               |              |                  |          |                                 |                          |     |
| CNC-impregnated |                           |            |               |              |                  |          |                                 |                          |     |

Note: The table includes data for various nanomaterials used in filtration membranes for wastewater treatment, including their thickness, porosity, flux, transmembrane pressure, permeability, and efficiency. Each entry represents a specific nanomaterial and its performance in filtrating different pollutants.
Table 7. Cont.

| Nano Celluloses | Membrane | Pollutants | Thickness (µm) | Porosity (%) | Flux (L.h⁻¹.m⁻²) | TMP (kPa) | Permeability (L.h⁻¹.m⁻².Pa⁻¹) | Efficiency (% or mg·g⁻¹) | Ref. |
|-----------------|----------|------------|----------------|--------------|------------------|----------|---------------------------------|--------------------------|-----|
| **Unmodified CNF** |          | Au         | 0.496-0.564    | 55.8-68.5    | 0.952-2.2        | -        | $1.19 \times 10^{-5}$-2.75 \times 10^{-5}$ | 84.6-93.5% [244] |     |
|                 |          | Ferritin (12 nm) | 0.496-0.564    | 55.8-68.5    | 0.952-2.2        | -        | $1.19 \times 10^{-5}$-2.75 \times 10^{-5}$ | 90.2-94.3% [244] |     |
|                 |          | Polyethylene glycol | 25-65          | 35           | -                | 200-10³  | $10^{-6}$-4 \times 10^{-6} | 88-100% [237] |     |
|                 |          | Polystyrene | 20-70          | 35           | -                | 200-10³  | $4 \times 10^{-6}$-2.7 \times 10^{-5}$ | 88-100% [80] |     |
|                 |          | Ag⁺         | 176            | 23-25        | 0-2.9            | 450      | 0-6.4 \times 10^{-6} | 77% [239] |     |
| Cellulose nanofibers | Oxidized CNF | Cu²⁺        | 176            | 23-25        | 0-2.9            | 450      | 0-6.4 \times 10^{-6} | 94% [239] |     |
|                 |          | Methylene blue | 0.496-0.564    | 55.8-68.5    | 0.952-2.2        | -        | $1.19 \times 10^{-5}$-2.75 \times 10^{-5}$ | 99.22% [244] |     |
|                 |          | Polyethylene glycol | 20-70          | 35           | -                | 200-10³  | $10^{-6}$-5 \times 10^{-6} | 58-100% [237] |     |
|                 |          | Polyethylene glycol Polystyrene | 20-70          | 35           | -                | 200-10³  | $1.2 \times 10^{-6}$-5 \times 10^{-6} | 58-95% [80] |     |
|                 |          | Ca²⁺/SO₄²⁻ | 20-70          | 35           | -                | 200-10³  | $10^{-6}$-4 \times 10^{-6} | 67-100% [80] |     |
|                 |          | Oil         | 20-100         | -            | 192              | 10³      | 1.92 \times 10^{-4} | 92.9-97.9% [217] |     |
|                 |          | Polyethylene glycol | 20-70          | 35           | -                | 200-10³  | $1.2 \times 10^{-4}$-2.0 \times 10^{-4} | 93% [247] |     |
|                 |          | White water effluent | 0.55-0.64      | -            | 98-122           | 500-10³  | $1.2 \times 10^{-4}$-2.0 \times 10^{-4} | 93% [247] |     |
|                 |          | Ag(I)       | 176            | 34-42        | 6.3-8.7          | 450      | $1.4 \times 10^{-5}$-1.9 \times 10^{-5} | - [239] |     |
|                 |          | Cr(VI)      | 200            | 78           | -                | -        | 0.145                       | 60% [222] |     |
|                 |          | Cr(VI)      | 40             | 80           | -                | -        | 0.189                       | 100% [247] |     |
|                 |          | Cu²⁺        | 25             | -            | -                | 200      | 2.3 \times 10^{-6} | 7.7% [245] |     |
|                 |          | Pb²⁺        | 176            | 34-42        | 6.3-8.7          | 450      | $1.4 \times 10^{-5}$-1.9 \times 10^{-5} | - [239] |     |
|                 |          | Acetone treated CNF | 200            | 78           | -                | -        | 0.145                       | 137.7 mg·g⁻¹ [222] |     |
| Nano Celluloses                        | Membrane                              | Pollutants | Thickness (µm) | Porosity (%) | Flux (L.h⁻¹.m⁻²) | TMP (kPa) | Permeability (L.h⁻¹.m⁻².Pa⁻¹) | Efficiency (% or mg·g⁻¹) | Ref. |
|---------------------------------------|----------------------------------------|------------|----------------|--------------|------------------|-----------|-------------------------------|---------------------------|-----|
| Thiol-functionalized CNF-impregnated  |                                        | Pb²⁺ Br⁻   | 40             | 80           | 215              | -         | 0.189                         | 260 mg·g⁻¹                | [247]|
| electrospun PAN                       |                                        |            |                |              |                  |           |                               |                           |     |
| Aminated CNF-impregnated electrospun |                                        |            |                |              |                  |           |                               |                           |     |
| PAN/PET                               |                                        |            |                |              |                  |           |                               |                           |     |
|                                        |                                        | Pb²⁺       | 40             | 80           | 215              | -         | 0.189                         | 260 mg·g⁻¹                | [247]|        |
|                                        |                                        | Br⁻        | -              | -            | 215              | -         | -                            | 89.6 mg·g⁻¹               | [248]|        |
|                                        |                                        |            |                |              |                  |           |                               |                           |     |
| Oxidized CNF-coated Ti-Bi oxide       |                                        | Cs⁺        | -              | -            | 215              | -         | -                            | 125.4 mg·g⁻¹              | [248]|        |
|                                        |                                        | I⁻         | -              | -            | 215              | -         | -                            | 225.9 mg·g⁻¹              | [248]|        |
|                                        |                                        | SeO₃²⁻     | -              | -            | 215              | -         | -                            | 204.5 mg·g⁻¹              | [248]|        |
|                                        |                                        | SeO₄²⁻     | -              | -            | 215              | -         | -                            | 85.1 mg·g⁻¹               | [248]|        |
|                                        |                                        | Sr²⁺       | -              | -            | 215              | -         | -                            | 81.4 mg·g⁻¹               | [248]|        |
|                                        |                                        | Polyethylene glycol | 16          | -            | 18–54            | 196–392   | 6.9 × 10⁻⁵–2.8 × 10⁻⁴         | 30–50%                    | [249]|        |
|                                        |                                        |            |                |              |                  |           |                               |                           |     |
| Bacterial cellulose (BC)              |                                        | Polyethylene glycol Oil | 15–85     | 35           | -                | 200–10³   | 5 × 10⁻⁶–2 × 10⁻⁵             | 60–75%                    | [237]|        |
|                                        |                                        |            |                |              |                  |           |                               |                           |     |
|                                        |                                        | Chlorella sp. | 20–100    | 95–97        | 441–845          | 10³       | 4.41 × 10⁻⁴–8.45 × 10⁻⁴       | 98.3–99.3%                | [7] |        |
|                                        |                                        |            |                |              |                  |           |                               |                           |     |
|                                        |                                        | Bovine serum albumin | 8–27       | 1.4–2.4      | 3.2–11.7         | 0–220     | 3.2 × 10⁻⁵–1.17 × 10⁻⁴        | 99.6–99.8%                | [249]|        |
|                                        |                                        |            |                |              |                  |           |                               |                           |     |
|                                        |                                        | Polyethenes serum glycol | 8–102     | -            | 0.03–6           | 196–392   | 7.6 × 10⁻⁸–1.5 × 10⁻⁵         | 90%                       | [236]|        |
|                                        |                                        |            |                |              |                  |           |                               |                           |     |
|                                        |                                        | BC derivative | Polyethylene glycol | 16–102 | -            | 8–15      | 196–392 | 5 × 10⁻⁶–5 × 10⁻⁵ | 85–90%                     | [236]|        |
|                                        |                                        |            |                |              |                  |           |                               |                           |     |
|                                        |                                        | Chitin laminated BC |             |              |                  |           |                               |                           |     |
|                                        |                                        | GO/BC      | 14             | -            | 1146             | -         | -                            | -                         | [250]|        |
|                                        |                                        |            |                |              |                  |           |                               |                           |     |
| Cobalt-phthalocyanine immobilized BC  |                                        |            |                |              |                  |           |                               |                           |     |
9. Water Disinfection Materials from Pathogenic Microorganisms

Disinfection processes are the last and most important step in the wastewater treatment process, i.e., removal, deactivation or killing of pathogenic microorganisms. Over the past decades, several nanomaterials (metal, metal oxides, and polymer NPs) have been used as disinfectants to reduce the harmful disinfection by-products [251]. Nanocelluloses on their own have negligible effects on microbial inhibition, and they can serve as a carbon source for their development, especially bacteria, algae, and fungi. Therefore, antimicrobial and antibacterial activities should be added to nanocellulose based materials to inhibit microbe’s growth in contaminated water. In this aspect, antibacterial features can be added to nanocelluloses for instance by surface modification with antimicrobial polymers, metal NPs (e.g., Ag, Au), and metal oxide NPs (e.g., TiO$_2$, CuO, ZnO, MgO) [252]. Surface modification of nanocelluloses with oxidized groups gives active sites (TEMPO oxidation) provides nanocelluloses with functionalized carboxyl groups with high affinity towards *E. coli* (rejection ~96–99%). The introduction of negative surface charges (carboxyl groups) offers resistance against Gram-positive and Gram-negative bacteria, and a high binding affinity to the negative charge near the bacterial membrane can be provided. The carboxyl groups on the nanocelluloses surface do not intrinsically act as antibacterial agents, but they can be loaded with appropriate antibacterial compounds. For instance, cationic surface modification by quaternary ammonium compounds, such as ammonium bromides [253], and ammonium chlorides [254], is a straightforward method to introduce antibacterial activity.

Nanocellulose also offers key functionalities that can be valued in antimicrobial filtration applications with antifouling properties. Nanocelluloses membranes with smaller pore sizes (~0.2 µm) in microfiltration membranes should allow the efficient elimination of bacteria (e.g., *E. coli*, *Salmonella*, *Hepatitis A virus*, *Cryptosporidium*). Mixtures of CNFs with cationically charged chitin nanofibrils allow the incorporation of antimicrobial activity while providing good stiffness and mechanical flexibility to the membranes [255]. In combination with the surface charge effect, the hydrophobicity variation through changes in the alkyl chain length enhances the compatibility with the lipid bilayer of the bacterial cells [256]. A study used antibacterial Ag NPs embedded in the matrix of an aluminum oxyhydroxide-nanocellulose composite to identify the optimum chloride concentration in water for effective antibacterial activity against *E. coli* and *B. subtilis*. The findings emphasized that increasing the chloride concentration drastically improves the antibacterial activity by reducing bacterial cell proliferation [257]. Recently, Nguyen et al., developed Ag NP-containing carboxylated CNF composites and dopamine (DA)-conjugated (CCNF-DA) with improved mechanical features, antimicrobial activity, and electrical conductivity by exploiting mussel’s utilization of catechols (Figure 13A). They found that the composite is anisotropically aligned due to the strong interaction between catechol moieties and Ag NPs, and that it endows electrically conductive biomaterials with flexibility, electronic conductivity, and antibacterial activity (Figure 13B). These materials may have a potential application as disinfectants for wastewater treatment.
10. Current Challenges and Limitations

Even though cellulose-based composite materials show promising application in water purification for the elimination of heavy metal ions and organic dyes, it remains a big challenge to control the main factors of porosity, permeability, and specific adsorption. The control over the morphology of the membranes can be controlled through the presented methods in this review (e.g., electrospinning, coating, casting, freeze-drying), but a continuous and most cost-efficient industrial process for fabrication of the membranes still follows an easy papermaking-like route. However, urgent limitations to be resolved are attributed to the energetic efficiency for the formation of nanocellulose membranes (e.g., fabrication of nanofibrils, dewatering of the fiber mats), quality of the produced membranes (e.g., the occurrence of defects in the membranes and homogeneity in pore sizes), and performance (e.g., the relatively thick membranes are required to compensate for defects while decreasing the permeability and increasing pressure drops). In particular, the economic and cost-efficient upscaling of production of suitable membranes needs to be developed, in particular, controlling film formation and drying procedures to improve the morphology of high-flux membranes. The strong tendency for agglomeration of nanocellulose during film drying can be resolved by the deposition of nanocellulose onto given filters or membrane structures providing them with a thin functional layer.

In parallel, the viable surface modification will remain crucial to improving the selectivity and anti-fouling properties. The native nanocellulose membranes show moderate efficacy for adsorption and photocatalytic degradation, and several issues must be overcome to synthesize composite materials with adequate pollutant removal efficiency. Pollutant adsorption can occur through chemisorption or hydrogen bonding that can be
facilitated by membrane surface functionalization with organic or inorganic materials. During adsorption, functional groups, such as $\text{−COOH}$ and $\text{−OH}$, strongly interact with the pollutant molecules and have important roles in inorganic mineralization and the immobilization of metal oxides [120]. Therefore, alternative processing routes should be developed enhancing the reproducibility of single nanofiber geometries and acquiring more impact on their agglomeration within a web-like structure with controlled porosity and surface properties through a one-step processing route. A limited number of processing steps should therefore be foreseen, likely by introducing controllable charges at the nanofiber surface while avoiding traditional wet chemistry and using green solvents.

The nanocellulose composite membranes have been proposed to increase the functionality (e.g., mechanical stability, chemical resistance, adsorption specificity, photodegradation) in particular for treatment of more complex water systems. A new aspect that will probably attract more attention in the future is the use of cellulose nanomaterials as the organic part in nanocomposites. The integrated cellulosic nanomaterial/inorganic material composites must be more thoroughly studied. More research is also required on cellulose/inorganic hybrids for the long-term treatment of natural water bodies and wastewater. However, the formation of cellulose/inorganic composite materials is a complex, not fully understood process. Several techniques have been used for the nanocomposite membrane synthesis, such as in situ precipitation [99], biomineralization [119], sol-gel [208], conventional casting [201], and co-precipitation [192]. The breakthrough of 3D printing can bring a more convenient method for the fabrication of complex nanocellulose composite membranes. Cellulose functionalization through oxidation or grafting improves its ability to initiate the growth of the inorganic nanoadsorbents and provide support for the immobilization of heavy metals [256]. One main challenge to the commercialization of nanocellulose composites is obtaining a homogenous mixture between cellulose and the inorganic nanoadsorbents. Indeed, homo-aggregation of the composite parts can negatively affect the morphology, efficiency and performance of the prepared composite materials [259]. Several techniques have been explored to address this issue, including the use of cellulose solvents [260], soluble cellulose derivatives [99], and inorganic mineralization control [261]. However, the regeneration and recyclability of the nanocomposite membranes should be considered from the design phase on.

11. Conclusions

Developing eco-friendly water treatment materials with improved performance has received significant academic and industrial attention. To date, a large variety of approaches have been developed to address arguably one of the key challenges for humanity in the 21st century: clean water. Among the different nanomaterials, nanocelluloses are promising sustainable materials for wastewater treatment, in terms of performance and efficiency, and also lightweight with strong mechanical strength, chemical inertness, and versatile surface chemistry, inexpensive production costs, and safe handling compared with inorganic nanomaterials. The improvement of nanocellulose production has led many researchers to fabricate nanocellulose-based materials (individual NPs, hydrogels, aerogels, sponges, membranes) for wastewater treatment. Different nanocellulose forms (cellulose nanocrystals, cellulose fibers, bacterial cellulose) have been successfully investigated for water treatment, particularly in membranes and filters (size exclusion, e.g., for nanoparticle filtration, or affinity membranes) and also as adsorbents (e.g., heavy metal ions, dyes, drugs, pesticides, fertilizers). These nanocellulose-based materials show good scalability and regenerative capacity, as indicated by their good performance after several cycles (adsorption-desorption). Nanocellulose functionalization by surface grafting or blending with different inorganic materials, polymers, and carbon materials allow the mass production of new renewable and sustainable cellulose composites at a lower cost. The unique features of nanocelluloses and their high compatibility with many modifiers make them appropriate for water treatment by adsorption, absorption, photocatalytic degradation, disinfection, antifouling, ultrafiltration, nanofiltration and reverse osmosis. However,
additional effort is required to implement and commercialize nanocelluloses as a viable nanomaterial for remediation technologies. Their dispersion stability and polydispersity, especially for industrial and commercial production, are another issue. The main challenge and limitations in working with nanocellulose-based materials must be identified to improve the development and efficient use of nanocelluloses in environmental remediation. The most effective strategy is to use nanocelluloses as support material for NP immobilization because NPs can freely penetrate the human body. However, impacts associated with a combination of nanocelluloses with functional NPs for use in wastewater treatment and the removal of NPs from the liquid after the reaction should be defined. This research field is rapidly growing and gearing towards the future commercial use of cellulose/inorganic hybrids for various industrial applications, including wastewater treatment.

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