Nanocellulose in Industrial Wastewater Treatment: An Overview

Vartika Srivastava

Abstract Over the past few decades, the urge for fresh and pure water has been rising each day. The need of the hour is to develop such viable technologies which are economical, have greater efficacy and lower carbon footprint over conventional methods. The current chapter features modern research studies related to the application of nanocellulose and its composites for the treatment of wastewater. Cellulose in the form of nanocrystal and nanofibrils are used effectively for the purpose of water purification owing to their unique properties. Nanocelluloses are bio-degradable, non-toxic materials used sustainably as nanofiller due to their remarkable mechanical properties, larger surface area, controllable surface chemistry and high aspect ratio. The chapter discusses the effectiveness of these materials for the removal of water pollutants through adsorption, catalytic degradation, photocatalysis, and flocculation. The mechanisms involved in the action of these processes are also discussed. Moreover, the limitations of these nanocellulose-based materials, along with the opportunities and the future prospects for wastewater treatment, have been discussed in detail.

1 Introduction

The evolution of Nanotechnology over the past few decades has redefined several industries and diminished the conventional scientific limitations. The objective of achieving viable development can be accomplished by substituting the nonrenewable sources of energy with biodegradable and green renewable sources around the world. In the past few years, Nanotechnology has come up as a budding approach for the removal of harmful pollutants such as greenhouse gases, high molecular weight organic compounds, toxic chemicals, and various biological agents. These nanomaterials are applied in several forms and morphologies (e.g., membranes,

V. Srivastava (\boxtimes)

Department of Basic Sciences and Humanities, Rajiv Gandhi Institute of Petroleum Technology, Jais 229304, India

e-mail: pc16001@rgipt.ac.in

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adsorbents or catalysts) for different functions. The high surface area of these nanomaterials imparts them higher reactivity and certain distinguished properties, which are advantageous for environmental remediation (Khin et al. [2012\)](#page-24-0). Currently, most of the research work for environmental remediation and energy production is focused on carbon and metal-based nanomaterials such as metal/metal oxide nanoparticles and nanocomposites, carbon nanodots (Cringoli et al. [2017\)](#page-23-0), two-dimensional carbonbased nanocomposites (Kumar et al. [2017;](#page-24-1) Chen et al. [2017;](#page-22-0) Ma et al. [2017a\)](#page-25-0). Although these materials exhibit excellent efficiency for environmental remediation, the major disadvantages associated with these materials are a requirement of non renewable sources for their production, toxic by-products, and biological incompatibility. The best possible substitute for these materials could be ones that are primarily abundant in nature. Considering the abundance of available resources of plants, plant extracts are the most reliable root for the fabrication of nanomaterials (Lu and Ozcan [2015\)](#page-24-2). Among the naturally available components, cellulose is the most abundantly found polymer, which has the ability to outgrow the persisting challenges related to cost, biodegradability, energy and renewability (Klemm et al. [2005\)](#page-24-3). The lignocellulosic biomass is extracted from fodder, i.e., agricultural waste and forest residues. These fodder crops are available in huge quantities, are extremely low-cost, and are also nugatory for human consumption, making them an even better choice over food crops as a renewable source for the fabrication of nanomaterials (Loow et al. [2017;](#page-24-4) Bhatnagar et al. [2015;](#page-22-1) Mahfoudhi and Boufi [2017\)](#page-25-1). Several articles are reported by researchers where nano cellulosic materials (CNs) have been applied for different applications such as paper making (Park et al. [2017a\)](#page-25-2), energy production (Du et al. [2017\)](#page-23-1), biomedical engineering (Liu et al. [2017\)](#page-24-5) and wastewater treatment (Putro et al. [2017\)](#page-26-0).

In this chapter, the application of nanocellulose for wastewater treatment has been discussed in detail. The two major forms of liquid waste that are most often discharged into the water bodies and causes harm to water ecology are wastewater and spilled oil. Industrial and municipal wastewater consists of an enormous amount of dissolved or suspended contaminants from different sources. Waste oil is spilled into the water bodies through different sources, including land drainage and disposal of waste. Moreover, the leakage from tank ship spills during their routine maintenance, and offshore oil production operation etc. also leads to spilling of oil in water bodies. The application of nanocellulose is highly effective for the removal of both types of liquid wastes (Mehmood et al. [2021\)](#page-25-3).

In view of all aspects, the aim of the chapter is to summarize the advanced methods for the preparation of nanocellulose-based composites that can be applied as biobased materials for the treatment of wastewater and also for remediation of wastewater. The nanocellulose-based materials are applied for wastewater treatment in the form of adsorbents, membranes, flocculants and also as catalyst carriers. Moreover, its shortcomings and perspectives on future research about further modifications in this area have been discussed.

2 Categorization of Nanocellulose

Nanocellulosic membrane can be majorly classified into two groups: (i) nano-objects and (ii) nanostructures (Kargarzadeh et al. [2017a\)](#page-24-6). These two groups are further classified into subgroups. Nano-objects are comprised of cellulose microcrystal and cellulose microfiber, while nanostructures are branched into cellulose nanocrystal (CNCs) and cellulose nanofibers (CNFs). The major difference between the two main groups is the size range of nano cellulosic materials. Nano-object cellulose is in the range of $10-100 \mu m$, while the size of nanostructured cellulose lies between 1 and 50 nm. Figure [1](#page-2-0) illustrates the length of CNF and CNC.

2.1 Cellulose Nanofiber (CNFs)

CNFs are identified by their specific structure having elementary nanofibrils alternating with crystalline and amorphous domains of cellulose (Kargarzadeh et al. [2017a\)](#page-24-6). Mechanical disintegration of the plant cell wall is done before turning the complex structure cellulose fiber into CNF. Depending on the power of disintegration, the diameter of fibers may range from 10–100 nm. Bacterial cellulose is the only type of nanocellulose fibers that are synthesized using enzymatic polymerization of organic compounds such as glycerol or sugar (Mahfoudhi and Boufi [2017;](#page-25-1) Gama et al. [2017\)](#page-23-2). Bacterial cellulose has exceptional purity, water holding capacity and crystallinity as compared to other types of CNFs (Mahfoudhi and Boufi [2017\)](#page-25-1).

Fig. 1 Size chart of nanocellulose-based on material length

2.2 Cellulose Nanocrystal (CNC)

CNCs are identified by their rod shape composed of crystalline regions separated from CNFs (Kargarzadeh et al. [2017a\)](#page-24-6). It is assembled up by a series of alternations starting with the isolation of the crystalline region by separating the amorphous region. This isolation is attained by applying acid digestion before other treatment methods. Overall, CNCs are obtained after multiple steps of purification and post **treatments**

3 Sources of Cellulose

Before the production of nanocellulose, it is important to identify the sources from where cellulosic materials can be extracted. There is a wide range of sources for obtaining cellulose, including algae, bacteria, plants and tunicates. Although under certain conditions, the final properties of CNF are mildly influenced by the source of cellulose, the properties of CNCs are entirely dependent on the source (Missoum et al. [2013;](#page-25-4) Smyth et al. [2017\)](#page-26-1). Thus, it is very important to select the suitable source and extraction technique for obtaining the nano cellulosic material of desired properties (García et al. [2016\)](#page-23-3). Moreover, the impact of the source, extraction and processing on the environment should also be taken into consideration (Rosa et al. [2010\)](#page-26-2).

3.1 Algae

The utilization of budding algal sources of green energy ensures environmental sustainability. The requirement of algae has been enhanced by executing researches on artificial photosynthesis such as $CO₂$ fixation and water splitting (Huang et al. [2018\)](#page-23-4). Algae is vitally used for the extraction of nanocellulose, which are exploited for the fabrication of adsorbents and thin films. Different varieties of algae, such as green algae (also known as *Cladophora*), are largely being explored by researchers for the production of nanocellulose. Green algae are found to be the most effective variety of algae when compared to others owing to their ability to resolve concerns related to water pollution in coastal areas. The study of the cell wall of various algae species exhibits that the *Cladophorales* possesses a highly crystalline cell wall (Nicolai et al. [1952\)](#page-25-5). As reported by Mihranyan, the higher crystallinity of the cell wall of *Cladophora* advocates its inert nature, which makes it insusceptible to chemical treatments when compared to cellulose derived from other conventional natural sources (Mihranyan [2011\)](#page-25-6). The nanocellulose derived from the plant source is used for the fabrication of membranes and adsorbents. The membranes prepared from *Cladophora* algae trap the particles of the swine influenza virus as effectively as industrial filters (Metreveli et al. [2014a\)](#page-25-7). Moreover, the beads prepared from green algal cellulose have the capacity to adsorb Pd (II) ions upto 80% within a time span of 2 h (Ruan et al. [2016\)](#page-26-3). The exploitation of algae for nanocellulose extraction is not only cost-effective but also helps adsorbs $CO₂$ gas, which reduces greenhouse gases (El-Safty et al. [2015\)](#page-23-5).

3.2 Bacteria

The scope of using microbial hosts is extensively being studied owing to the multifunctional benefits derived from nanocellulose materials. Similar to algal cellulose, bacterial cellulose exhibits high purity, which is free from other polymers or functional groups except for the alcoholic group (Nechyporchuk et al. [2016;](#page-25-8) Uzyol and Saçan [2017\)](#page-26-4). The smaller degree of purification, which is required, is also less energy consumption as per the research conducted by Gatenholm and Klemm [\(2010\)](#page-23-6), the time required for the production of cellulose through bacteria is up to two weeks. Bacterial celluloses are prepared through the process of fermentation, wherein the movement of bacteria is either free in the culture medium or sometimes get attached to cellulose fiber forming swollen gel structure. These swollen balls are then purified by the death of the microorganism followed by the removal of cell waste and culture medium. The cellulose produced by bacteria is necessarily nanocellulose as fibrils can be united by bacteria only at the nanoscale. The nanocellulose is produced by different types of bacterial species such as *Acetobacter xylinum* (presently *Gluconacetobacter xylinus* or *Komagataeibacter medellinensis*) and *Gluconacetobacter medellinensis* (Gatenholm and Klemm [2010\)](#page-23-6). The production of cellulose from these species occurs by polymerization of monomeric units of cellulose, i.e., glucose, into the external environment. This complex process leads to the formation of a three-dimensional network of microfibril and nanofibril. This 3D network imparts crystallinity, mechanical strength, and water retaining capacity to the nanocellulose.

3.3 Plants

The major source of nanocellulose which is studied across the board is fabrication using plants. Plant nanocellulose can be majorly categorized into two types: (i) cellulose nanofibers and (ii) cellulose nanocrystals (Kargarzadeh et al. [2017b\)](#page-24-7). Plant nanocellulose can be obtained from a variety of sources (Ventura-Cruz and Tecante [2019\)](#page-26-5), such as coconut husk fiber, corncob residue, barley wastes, mengkuang leaves (*Pandanus tectorius*), garlic straw residues, raw cotton linter, *Agave tequilana,* tomato peels, forest residues, *Gigantochloa scortechinni* bamboo culms, industrial waste cotton, cassava root bagasse and peelings, sugar palm fibers (*Arenga pinnata*), corn straw and sago seed shells. Plant fibers used as a source of cellulose are classified into several groups, including bast fiber, grass fiber, core fiber, reed fiber, leaf

fiber, seed fiber and other fibers. Among all these, the most frequently used plant fiber for nanocellulose production is wood pulp. Wood pulp has a relatively higher content of pure cellulose, ductility and durability when compared to other plant sources (Menon et al. [2017\)](#page-25-9). Wood pulp nanocellulose has been reported to be used effectively used as a microfiltration membrane for the removal of bacteria, viruses and heavy metal ions from water bodies. In addition to the membrane, wood pulp nanocellulose has also been used to design catalysts for reducing hazardous organic compounds like 4-nitrophenol. Similarly, bleached birch fibres obtained from *Betula verrucosa* and *Betula pendula* have been used by Suopajärvi et al. [\(2013\)](#page-26-6) for fabrication of dicarboxylic acid nanocellulose applied for the treatment of wastewater. On account of these facts, plant based nanocellulose is considered the most beneficial and environment friendly source for the production of nanocellulose.

3.4 Tunicates

Tunicates (marine invertebrate animals) are the contemporary alternative for the fabrication of nanocellulose. Cellulose composed of highly crystalline and pure triclinic (CI_6) allomorphs is generally being deposited on the outer issue of the tunicates (such as sea squirts) (Kargarzadeh et al. [2017a\)](#page-24-6). Researchers have explored different species of sea squirts, including *Metandroxarpa ueda* (Kimura and Itoh [1996\)](#page-24-8), *Halocynthia roretzi* (Elazzouzi-Hafraoui et al. [2008\)](#page-23-7) and *Halocynthia papillosa* (Iwamoto et al. [2011\)](#page-23-8), for the production of cellulose microfibrills. Cellulose derived from tunicates has been applied in several works for environmental remediation. A study reported by Cheng et al. [\(2017\)](#page-22-2) discussed the synthesis of a filter membrane designed by homogenizing the nanocrystals of tunicate cellulose which are hydrophilic in nature with cholesteric liquid crystals. These filter membranes were found to be highly efficient for the separation of oil/water. Moreover, stalked sea squirts (*Styela Clava*) have also been used for the fabrication of nanocomposites. These nanocomposites have been effectively used as catalysts in the process of water treatment (Wei et al. [2014\)](#page-27-0). In research carried out by Yu et al. [\(2016a\)](#page-27-1), nanocomposites derived from stalked sea squirt were used as flocculant for flocculating and harvesting microalgae used for the production of biodiesel.

4 Methods of Preparation of Nanocellulose

4.1 Mechanical Disintegration

The most accepted method for the preparation of CNF is the mechanical disintegration used as a principal treatment to generate CNFs (Carpenter et al. [2015\)](#page-22-3). In

the process of fabrication of CNFs, mechanical disintegration is executed as a posttreatment step used to serve the purpose of purification (García et al. [2016\)](#page-23-3). This method is applied for breaking down the heavier chunks of cellulose pulp into smaller particles. For an efficient mechanical disintegration to take place, delamination of cellulosic nanofibrils is required instead of basic fiber shredding (Nechyporchuk et al. [2016\)](#page-25-8). The mechanical disintegration of cellulose pulp, which is subjected to fiber shredding, produces nanocellulose possessing poor mechanical properties. For enhanced delamination of nanofibrils, interfirillar hydrogen bonding needs to be loosened for inhibiting the fibril aggregation. This condition is satisfied by employing an aqueous medium during the process of mechanical disintegration. The delamination of cellulose fibers is carried out by using techniques such as homogenization (Abdul Khalil et al. [2014\)](#page-22-4), refining (Sacui et al. [2014\)](#page-26-7) and grinding (Nair et al. [2014;](#page-25-10) He et al. [2018\)](#page-23-9). The nanocelluloses generated using mechanical disintegration methods are of larger dimensions and heavily clustered (Sacui et al. [2014\)](#page-26-7). Park et al. [\(2017b\)](#page-25-11) reported that the defibrillation efficiency of wood based CNF in wet-disk milling is influenced by its chemical composition. The defibrillation efficiency is improved in the absence of lignin and hemicelluloses in the source material. The curtailment of viscosity, crystallinity and thermal stability conveys the transition of materials from micro to nanosize. In the majority of the cases, the most prominent mechanical treatments used include blending, electrospinning, grinding, ball milling, homogenization, cryocrushing, refining, ultrasonication, steam explosion or a combination thereof.

4.2 Chemical Method

The demand for large scale production of nanocellulose can be fulfilled by applying preparation techniques that are effective and low energy consuming. The chemical treatment of cellulose has been experimented with by researchers for the generation of nanocellulose. Fan et al. (2011) reported that the defibrillation process used for the production of nanocellulsoe is executed effectively by involving chemical agents (variation in pH). Chemical pretreatment techniques which are widely used in the current scenario include acid hydrolysis (Vanderfleet et al. [2018\)](#page-26-8), sulphonation (Rocha et al. [2018\)](#page-26-9), carboxylation (Sharma et al. [2018\)](#page-26-10), quaternization (Santos et al. [2018\)](#page-23-11), carboxymethylation (Onyianta et al. [2018\)](#page-25-12), ionic liquid (Ninomiya et al. [2018\)](#page-25-13) and pretreatments assisted by solvents (Laitinen et al. [2017\)](#page-24-9). The most common technique for extracting CNC from cellulose is acid hydrolysis. The technique is helpful for destroying the non-crystalline region present in the microfibril. The destruction of the amorphous region helps in keeping the crystalline portion intact. Liu et al. demonstrated the fabrication of CNC through hydrolysis of sulphuric acid (Liu et al. [2014\)](#page-24-10). Other than CNC extraction, bacteria nanocrystals can also be generated from bacterial microfibrils via acid hydrolysis (Börjesson and Westman [2015\)](#page-22-5). Chemical treatment is used as a pre-treatment technique before the process of mechanical disintegration for preparing nanocellulose. These techniques are used to bring in

the charge on the surface of the cellulose. The modification in the surface charge by sulphonation, carboxylation, and carboxymethylation, assists in the defibrillation of cellulose fibers (Nechyporchuk et al. [2016\)](#page-25-8). Wågberg et al. [\(2008\)](#page-26-11) prepared thin nanofibrils through carboxymethylation of CNFs. The nanofibrils fabricated with chemical pre-treatment exhibited uniform width distribution and minimum agglomeration when compared to those prepared directly through mechanical disintegration without pre-treatment.

4.3 Biological Method

The biological method is an effective method for the hydrolysis of chemical waste. Enzymatic hydrolysis is a pre-treatment step which assists in combating issues related to energy cost and environmental hazards. This pre-treatment step is applied before mechanically disintegrating cellulose to nanocellulose through methods like refining or blending. The delamination of the cell wall during the disintegration of cellulose is further enhanced by the incorporation of the endoglucanase enzyme in a homogenizer (Pääkkö et al. [2007\)](#page-25-14). The average molar mass and aspect ratio of nanofibers manufactured through this treatment were found to be greater than those nanofibers which are fabricated through chemical treatment involving acid hydrolysis (Henriksson et al. [2007\)](#page-23-12). Besides endoglucanase, various other enzymes such as cellulases (Beltramino et al. [2015\)](#page-22-6), ligninases, xylanases (Tibolla et al. [2014\)](#page-26-12), pectinases (Hideno et al. [2014\)](#page-23-13) etc., have also been used for enzymatic hydrolysis of cellulose to nanocellulose.

Beltramino, along with the co-workers, synthesized CNC crystals that have superior dimensions and a lesser quantity of sulphur when compared to cotton linter (Beltramino et al. [2015\)](#page-22-6). This CNC was generated using the enzyme substrate of cellulose accompanied with acetate buffer. On the contrary, banana fibers obtained through chemical treatment have higher crystallinity in comparison to those obtained through xylanase treatment. The reason for this lower crystallinity is that these enzymes are not easily soluble in hemicelluloses which make it difficult for them to penetrate the cellulosic chains to hydrolyze them. The presence of hemicelluloses in the cellulose sources clogs the separation of cellulose nanofibrils. Nechyporchuk et al. [\(2016\)](#page-25-8) studied the rheological behavior of CNF produced with both enzymatic and chemical treatment. It was found that the flocculation capacity of CNF generated via enzymatic treatment is better than that obtained by chemical treatment. Figure [2](#page-8-0) represents the method of preparation of nanocellulosic materials.

5 Nanocellulose Materials Used for Water Treatment

The deteriorating quality of water resources is a serious matter of concern all over the globe. The perpetually growing urbanization and industrialization have led to

Fig. 2 Sources of cellulose and methods of preparation of CNCs and CNFs

an increase in the release of toxic substances into the water bodies. The decontamination of water bodies is an urgent need of the hour. Of lately, a lot of work is being done on utilizing environment friendly and long-lasting products derived from natural resources for their potential application in wastewater treatment. The environment friendly cellulose material in the micro and nanosize range is advantageous over conventional cellulose fibers owing to their high surface area, aspect ratio, and Youngs' modulus (Suopajärvi et al. [2014\)](#page-26-13). These cellulose-based nanomaterials are supremely versatile and have found their applications for various purposes. In this chapter, a summary of contemporary applications of cellulose-based materials as adsorbents, photocatalysts, flocculants and membranes for wastewater treatment has been reviewed and is represented in Fig. [3.](#page-9-0)

5.1 Nanocellulose-Based Adsorbents

The purification of wastewater using adsorbents derived from nanocellulose is currently a promising method in contrast to high energy consumption and overpriced technology employing carbon-based adsorbents. Unaltered cellulose does not possess the desired adsorptive property, therefore tailoring of cellulose is needed to achieve improved interaction and a productive outcome. The vital properties acquired by cellulose are (i) its hydrophilic nature, (ii) its ability to functionalize, (iii) its

Fig. 3 Schematic diagram describing the various water/wastewater treatment processes in which CN based systems can be used

tendency to reconcile its properties such as surface area, quantum size, aspect ratio and chemical accessibility (Anirudhan and Rejeena [2012\)](#page-22-7).

5.1.1 Adsorption of Heavy Metal Ions

The utilization of heavy metal ions in different industries poses a serious threat to human health. Metal ions used in industries (such as copper, silver, chromium, iron etc.) are leaked into the water bodies, including groundwater and contaminate these water sources. The intake of these ions by a human being in excess would have threatening repercussions (Bilal et al. [2013\)](#page-22-8). Jalali and Aboulghazi et al. [\(2013\)](#page-24-11) and Johari et al. [\(2016\)](#page-24-12) used plant-derived cellulose as an adsorbent for metal ions. Cellulosederived from sunflower stalk and coconut husk waste was used for adsorption of Cd (II) and Pb (II) metal ions and Hg (II) metal ions, respectively. In contrast to this, Bediako et al. [\(2016\)](#page-22-9) prepared carboxymethyl cellulose using lyocell fabric via crosslinking reactions followed by carboxymethylation. The carboxymethyl cellulose was used to adsorb Cd (II) ions from their aqueous solution. The process of adsorption was found to be approximately 17 times more efficient. The efficiency of adsorption of Cd (II) using nanofiber is notable even at a lower concentration. Sun et al. [\(2017\)](#page-26-14) prepared a cellulose adsorbent by halogenation of microcrystalline cellulose. Halogenated cellulose was then functionalized with pyridine diacid and further used for the removal of Pb (II) and Co (II). Several other chemicals have

been reported by researchers for modification of CNCs for making them an efficient adsorbent (de Castro Silva et al. [2018;](#page-22-10) Jain et al. [2017;](#page-24-13) Li et al. [2019;](#page-24-14) Madivoli et al. [2016;](#page-25-15) Mautner et al. [1861;](#page-25-16) Sun et al. [2018\)](#page-26-15). Rafieian et al. [\(2019\)](#page-26-16) made an attempt to prepare and characterize a CNC membrane containing (3-aminopropyl) triethoxysilane or APTES (MCNC) and a non-woven substrate at the bottom for removing Cu (II) ions in water. It was observed that the efficiency of removal of metal ions was enhanced using polyethersulfone (PES)/MCNC membrane. MCNC has the ability to upgrade the fouling resistance and water flux of the membrane. Similarly, Mathew and co-workers (Liu et al. [2014,](#page-24-10) [2015\)](#page-24-15) studied the potential of pristine surface functionalization of CNCs for adsorption of metal ions such as $Ag (I)$, Cu (II), and Fe (III) from water bodies. A study of both the classes of CNs, i.e. CNCs and CNFs derived from cellulose sludge, was carried out for adsorption of Ag (I) ions. CNCs were prepared by acid hydrolysis while CNFs were homogenization of cellulosic fibers. The surface area of fabricated CNCs and CNFs were found to be $138-226$ m²/g and $146-219$ m²/g, respectively. Adsorption experiments were carried out over a range of pH. It was found that adsorption of Ag (I) ions make the CNC suspension stable, whereas no stability was seen in Ag (I) treated CNF suspension. CNF suspensions were mostly sedimented, and well-defined layers were separated from them owing to the surface charge characteristics of CN/Ag (I) complexes. Moreover, the maximum adsorption capacity for CNCs and CNFs were calculated to be 0.32 and 0.14 mmol/g, respectively. From these observations, it can be concluded that CNCs do not settle down in solution even after treatment with Ag (I) ions and have higher adsorption

5.1.2 Adsorption of Dyes

capacity due to larger surface area.

The chemical discharge from several industries, including textile, pharmaceutical, leather tanning, pulp and paper, comprises different types of dyes that pollute the water bodies and needs to be removed necessarily (Crini [2006;](#page-23-14) Rafatullah et al. [2010\)](#page-26-17). Lately, several articles have been reported on the development of CNs-based adsorbents for the removal of these dyes from water bodies. CNCs obtained from acid hydrolysis were examined by Batmaz et al. [\(2014\)](#page-22-11) for their potential to attract positively charged dye molecules such as methylene blue. The adsorption capacity was found to be good enough, which was further upgraded by modification of these CNCs by functionalization via 2, 2, 6, 6-tetramethylpiperidin-1-oxyl (TEMPO) oxidation. It was found that the adsorption capacity was improved by up to 6 times. The CNCs developed by acid hydrolysis possess a negative charge on its surface, and oxidation of its primary hydroxyl groups using TEMPO reagents introduced more negative charges. The increase in negative charge facilitated its easier interaction with positively charged dye molecules. The maximum adsorption capacity of unaltered CNCs and TEMPO-oxidized CNCs were found to be 0.37 and 2.40 mmol/g, respectively. The increase in the value of maximum adsorption capacity indicates the adsorption of positively charged dye molecules over anionic sites of adsorbent as a result of counter ion exchange.

Gholami Derami et al. [\(2019\)](#page-23-15) used thin films composed of nanocellulose with polydopamine (PDA) for adsorbing toxic dyes such as rhodamine 6G, methylene blue and methyl orange.

Apart from targeting cationic dyes, CNCs are also modified to act as an adsorbent for anionic dyes from their aqueous solutions. Eyley and Thielemans [\(2011\)](#page-23-16) grafted the imidazolium group over CNCs through azide-alkyne cycloaddition reaction catalyzed using a heterogeneous Cu (I). The cycloaddition of imidazolium groups imparts positive charges over grafted CNCs. These positively charged species were capable of attracting negatively charged dye molecules such as orange II. The maximum adsorption capacity of this adsorption process was found to be 0.28 mmol/g.

Apart from the grafting of these nanocellulose membranes, double-layered membranes are also being prepared by implementing a layer of some other material over the CNF membrane. It was observed that merging the CNF membrane with that of graphene oxide improves the adsorption efficiency of the membrane (Liu et al. [2019\)](#page-24-16). The merged membranes produce synergistic membranes owing to the presence of inter and intramolecular hydrogen bonds forming a network (Sajab et al. [2016;](#page-26-18) Fang et al. [2016\)](#page-23-17). These membranes were used for the removal of dyes such as rhodamine 6G, victoria blue and methyl violet from their aqueous solution.

5.1.3 Adsorption of Other Water Contaminants

Besides dyes, oils and heavy metal ions, there are several other substances which may contaminate water bodies. The presence of pharmaceuticals, agrochemicals, and biomolecules in municipal/industrial wastewater may create major health problems. Researchers have made attempts to evolve CN based materials to remove these contaminants. Chen et al. (2014) developed β-cyclodextrin modified $CNCs@Fe₃O₄@SiO₂ superparamagnetic nanorods. These nanorods were applied$ for the adsorption of pharmaceutical wastes such as procaine hydrochloride and imipramine hydrochloride. These pharmaceutical compounds got trapped into the hydrophobic cavity of β-cyclodextrin grafted on the CNCs $@Fe_3O_4@SiO_2$ superparamagnetic nanorods. After the completion of the adsorption process, these nanoparticles could be easily recovered from the aqueous solution under the influence of a magnetic field. The maximum adsorption capacity for procaine hydrochloride and imipramine hydrochloride was found to be 0.055 and 0.052 mmol/g, respectively. In a study reported by Herrara-Morales et al. [\(2017\)](#page-23-18), CNC composites were altered by polyethylene glycol (PEG). These composites were utilized for the separation of pharmaceutical compounds such as acetaminophen, sulfamethoxazole and N, N-diethyl-meta-toluamide (DEET) from aqueous solutions. PEG provides ease in immobilization of poorly soluble drugs in water which activates the interaction between CNCs and hydrophobic drugs. The modified composite acquired improved physical properties such as high surface area and hydrophilicity, which permitted the adsorption of pharmaceutical compounds via electrostatic interactions.

Similar to pharmaceutical compounds, insecticides such as chlorpyrifos was also targeted by Moradeeya et al. [\(2017\)](#page-25-17) for their removal from wastewater using pristine CNCs. The maximum adsorption experiment for this adsorption experiment which takes about 60 min to reach equilibrium, was found to be 0.027 mmol/g. It was also reported by them that using an eluent solution comprising of 80% methanol and 20% water could completely separate chloropyrifos from CNC during the cycle of adsorption–desorption. Researchers have also shown the budding approach of CN based systems for the adsorption of biomolecules. CNCs functionalized with di-aldehyde were prepared by Huang et al. [\(2016\)](#page-23-19) for the adsorption of creatinine. Di-aldehyde groups were attached to the surface of CNCs through sodium peroxide oxidation (referred to as D-CNCs). The increase in aldehyde content increased the adsorption capacity of D-CNCs, while the reduction in the size of D-CNCs enhanced the rate of reaction. The maximum adsorption capacity of D-CNCs having the highest aldehyde content was calculated to be 0.013 mmol/g. The mechanism of adsorption of creatinine is the crosslinking reaction taking place between the aldehyde group of D-CNCs and the amino group of the creatinine molecule. Anirudhan and co-workers targeted (Anirudhan and Rejeena [2012,](#page-22-7) [2013a,](#page-22-13) [b\)](#page-22-14) adsorption of biomolecule known as trypsin using CNCs modified with carboxylate functionalized cation exchanger such as polyacrylic acid modified polyglycidylmethacrylate attached to CNCs. The maximum adsorption capacity for this adsorption process that attains equilibrium within 90 min was found to be 0.006 mmol/g. The adsorbent used in a cycle is 100% regenerated by using 0.1 mol/L KSCN as eluent (Anirudhan and Rejeena [2012\)](#page-22-7). Besides this, a novel adsorbent for the removal of proteins such as immunoglobulin (IgG) and haemoglobin (Hb) was also developed by grafting magnetic CNC composites with poly (methacrylic acid-co-vinyl sulfonic acid). The maximum adsorption capacity for adsorption of Hb and IgG attaining equilibrium in 2 and 3 h were found to be 0.003 and 0.013 mmol/g, respectively. This novel adsorbent selectively removes the Hb or IgG from their mixture with bovine serum albumin. The adsorption takes place under the influence of a magnetic field owing to the magnetic properties of the modified CNCs associated with $Fe₃O₄$. Regeneration of adsorbent is carried out using 0.01 mol/L KOH eluent without compromising on its adsorptive capacity (Anirudhan and Rejeena [2013a,](#page-22-13) [b\)](#page-22-14). In each of these reports, biomolecules get adsorbed via electrostatic attraction between the adsorbent possessing a negative charge and positively charged protein molecule below their isoelectric point. The detailed list of CN-based adsorbents used for heavy metal ions, insecticides, anions and proteins is given in Table [1.](#page-13-0)

5.2 Oil–Water Separation

Nanofibrillated cellulose is also used to prepare nanocomposites in the form of aerogels (Mehmood et al. [2021\)](#page-25-3). These aerogels possess a structure having a strong network through interfibrillar hydrogen bonding via the freezing–thawing process. This nenofibrillated cellulose (NFC) aerogels are effectively used as oil sorbents. In

Cellulose based adsorbent	Heavy metal ions	Maximum adsorption capacity (mmol/g)	References
Sulphonated nanofibrillar cellulose	Pb(II)	248.6	Suopajärvi et al. (2015)
Pristine CNCs	Ag(I) Cu (II) Fe (III)	0.52 0.31 0.11	Liu et al. (2015)
Succinic anhydride modified CNCs	Pb(II) Cd (II)	1.77 2.31	Yu et al. (2013)
APTES modified CNFs	Ni (II) Cu (II) Cd (II)	2.73 3.15 4.19	Hokkanen et al. $(2014a)$
Poly (itaconic acid)-poly (methacrylic acid)-grafted-nanocellulose/nanobentonite composite	Co (II) U(VI)	350.8 121.02	Anirudhan et al. (2016)
Pristine CNCs	Methylene blue	0.37	Batmaz et al. (2014)
Maleic anhydride grafted CNCs	Crystal violet	0.59	Qiao et al. (2015)
Imidazolium grafted CNCs	Orange II	0.28	Eyley and Thielemans (2011)
Functionalization of cellulose with hyperbranched polyethylenimine	Congo red Basic yellow 28	2100 1860	Zhu et al. (2016)
MnO ₂ coated CNFs	Methylene blue	$\overline{}$	Wang et al. (2014a)
Pristine CNCs	Chlorpyrifos	0.027	Moradeeya et al. (2017)
D-ialdehyde functionalized CNCs	Creatinine	0.013	Huang et al. (2016)
APTES modified CNFs Epoxy modified CNFs	Hydrogen sulphide	3.05 0.373	Hokkanen et al. (2014b)
Cationic CNFs functionalized with glycidyltrimethylammonium chloride	$PO4$ ³⁻ $SO_4{}^{2-}$ F^- NO ₃	55 50 10.6 44	Sehaqui et al. (2016)
Poly (methacrylic acid-co-vinyl sulfonic acid) grafted magnetic CNCs	Haemoglobin (Hb) Immunoglobulin (IgG)	0.003 0.013	Anirudhan and Rejeena (2013a, b)

Table 1 CN-based adsorbents used for removal of water pollutants

CNC based adsorbent	Adsorbent characteristics	Adsorption capacity	References
Cellulosic scaffolds made from acetate esterified CNCs	$\rho = 3.3$ to 7.5 kg/m ³ ; θ $= 140^\circ$	$100 - 200$ g/g ^a	Anirudhan and Deepa (2017)
CNF aerogels treated with methyltrimethoxysilane	$\rho = 5.07$ to 17.3 mg/cm ³ ; Porosity = $>99\%$, SSA $=$ 3 to 25 m2/g, θ = 110 to 150°	49-102 g/g^a	Zhang et al. (2014)
CNF aerogels coated with $TiO2$	$\rho = 20$ to 30 mg/cm3; Porosity = $>98\%$, θ = $>90^\circ$	$20 - 40$ g/g ^a	Korhonen et al. (2011)
Hydrazone-carboxyl ligand-linked CNC based aerogel	$\rho = 22.4$ to 23.3 mg/cm ³ ; Porosity 96.3–97.2% $SSA = 195$ to 303	133 g/g (for water), 99 g/g (for ethanol), 34 g/g (for toluene) 54 g/g (for dodecane)	Ma et al. (2017b)

Table 2 Oil/water separation using various CN based adsorbents

^a Adsorption capacity for a range of organic solvents and oils, $ρ$ -density, SSA–specific surface area, θ-contact angle

this regard, Gao et al. [\(2018\)](#page-23-22) prepared superhydrophobic NFC aerogels by firstly coating the NFC with dopamine/octadecyl amine (ODA) accompanied by a freeze drying process. This leads to the formation of a PDA coating acting as a layer between the NFC platform and ODA. The fabricated aerogel possessed a high contact angle of 152.5 \degree and an ultralow density of 6.04 mg/cm³, which provided buoyancy to these materials and aided them with exceptional oil/water absorption selectivity. Similarly, Chhajed et al. [\(2019\)](#page-23-23) prepared NFC/PVA (polyvinyl alcohol) aerogels with the coating of stearic acid chloride (SAC). These SAC conjugated aerogels also showed superhydrophobicity and a contact angle of 0° . The capacity of adsorption is gauged by the ratio of the weight of liquid adsorbed to the dry weight of aerogel. NFC/PVA aerogels showed remarkable selectivity in the separation of oil/water mixtures along with other organic solvents. Table [2](#page-14-0) represents the list of cellulose based adsorbents used for oil/water separation.

5.3 Nanocellulose-Based Photocatalyst

Recently, a lot of work has been done on employing several inorganic–organic hybrid based nanocomposites possessing distinct electrical, thermal and mechanical properties for photocatalysis (Yang et al. [2011\)](#page-27-6). The transition from petrochemical-based feedstock to biomaterials is slowly making its way for a sustainable and cleaner ecosystem. Cellulose acts as biocompatible support due to the chirality, broad chemical variability, and hydrophilicity possessed by this material. Although cellulose alone is not a good photocatalytic agent, the incorporation of semiconductor materials into its matrix enhances its photocatalytic activity by several folds. Research

Cellulose based photocatalyst	Model pollutant	Degradation efficiency $(\%)$ and reaction time (min)	References
CdS nanoparticle/bacterial cellulose nanofibers	$200 \text{ mL} 20 \text{ ppm}$ methyl orange	82%, 90 min	Yang et al. (2011)
N -doped TiO ₂ nanorods in regenerated cellulose thin films	150 mL 40 mg/L methylene blue	96%, 360 min	Mohamed et al. (2015)
Graphene oxide/TiO ₂ based ultrafiltration cellulose membranes	$3.40 \times 10 - 5$ mol/L diphenhydramine at flow rate of ≈ 0.25 mL/min	65%, 240 min	Pastrana-Martínez et al. (2015)
Alginate/carboxymethyl cellulose/ $TiO2$ nanocomposite hydrogel	30 mL 30 mg/L Congo red	91.5%, 240 min	Thomas et al. (2016)

Table 3 CN-based hybrid photocatalysts for degradation of organic pollutants

groups are now working on examining cellulose-based metal oxide nanomaterials for photocatalytic wastewater treatment in the form of membranes, fibers, thin films and hybrid materials under UV and visible light irradiation (Table [6\)](#page-20-0). Metal oxides have the ability to adsorb cellulose on their surface, which creates additional –OH groups on their surface. In addition to the –OH group, the fibrous network also assists the adsorption of cellulose on the surface of metal oxide. Li et al. [\(2017\)](#page-24-18) studied the incorporation of $TiO₂$ nanoparticles within the fibrous network matrix of bacterial cellulose via hydrogen bonds. The modified cellulose material was used for photocatalytic degradation of reactive $X-3B$. TiO₂ nanoparticles were bounded with cellulose chains through a covalent bond which made the polymer chains rigid. The improved rigidity of the polymer chains enhanced the stability of these chains (Mohamed et al. [2015\)](#page-25-19). Similar results were obtained with CdS nanocrystals coated on cellulose derived from microbes (Yang et al. [2011\)](#page-27-6). The nanocomposite catalyst showed excellent photocatalytic activity and successfully degraded 82% of dye in 90 min. of time span. These catalysts were 100% recovered after each cycle without losing their efficiency. A detailed list of cellulose-based photocatalysts is given in Table [3.](#page-15-0)

5.4 Nanocellulose-Based Catalyst Carriers

In addition to photocatalysis, fibrillated cellulose also has the potential to act as a support/ carrier for nanoparticles used as a catalyst. The porous structure and presence of a large number of functional groups on their surface make these materials an ideal catalyst carrier. An et al. [\(2016\)](#page-22-17) studied the use of nanocellulose as support for $TiO₂$ nanoparticles to fabricate nano-fibrillated-TiO₂ nanoparticles nanocomposite

for hydrogen generation through photocatalysis. The capacity of hydrogen generation by nanocomposites was found to be greater than that of $TiO₂$ nanoparticles. However, it was found that nanocomposites photo degraded during the course of the photocatalytic reaction and hence, 100% catalyst could not be generated at the end of a cycle. Another study reported by Ren et al. [\(2018\)](#page-26-23) discussed the preparation of metal–organic frameworks (MOFs)/cellulose aerogels used as a catalyst for the removal of organic pollutants, including rhodamine B 4-nitrophenol and tetracycline hydrochloride. The MOFs were applied for triggering peroxymonosulphate (PNP) to react with the above mentioned contaminants. After the end of the reaction, MOFs were separated from the solution by using zeolite imidazole framework (ZIF) materials (ZIF-9 and ZIF-12). Cellulose aerogels were soaked into the solution of ZIF materials to load the ZIF materials (ZIF-9 and ZIF-12) on cellulose aerogels. The degradation of PNP was then carried out. The experiments showed that a hybrid aerogel system could degrade 90% PNP in 1 h. The aerogels were also regenerated easily from the solution. The efficient degradation of PNP showed the budding prospects of applying hybrid aerogels as a catalyst in the advanced oxidation process. In another study carried out by Taranto et al. [\(2009\)](#page-26-24), it was observed that cellulose exhibited sensitivity towards UV radiation and, therefore, is susceptible to degradation during photocatalysis. Degradation of cellulose is accompanied by degradation of other organic contaminants such as methanol producing photocatalytically active species. The degradation of cellulose might compete with organic contaminants leading to a reduction in the rate of reaction. Moreover, Puls et al. (2011) also explained that under the illumination of UV light, TiO₂ nanoparticles could create holes on the surface of cellulose acetate/cellulose pulp fibers. Therefore, to prevent UV bleaching, coating of the cellulose nanomaterials need to be done for covering the holes perfectly by inert materials capable of absorbing UV light. Table [4](#page-17-0) represents such reports where nanocellulose has been used as a catalyst carrier.

5.5 Nanocellulose-Based Flocculants

Flocculation is a technique used for removing fine solid particles of metals, dyes and other organic compounds suspended in water bodies. The addition of flocculants accelerates the colloidal particles to collide among themselves and form bigger unstable particles. These bigger particles, known as flocs, precipitate from the solution in due course of time (Lee et al. [2014;](#page-24-19) Quinlan et al. [2015\)](#page-26-26). In recent times, majorly used flocculants include inorganic, cationic and multivalent coagulants such as aluminium iron salts and synthetic polymers like polyacrylic acids and polyacrylamides derived from petroleum-based non-renewable sources are applied as flocculants in wastewater treatment (Suopajärvi et al. [2017\)](#page-26-27). The use of these flocculants is hazardous to the environment. Therefore, research efforts are being made to substitute these harmful substances with biodegradable, biocompatible and long lasting flocculants based on CNs.

Cellulose based catalyst	Catalytic conversion	Conversion efficiency	References
Cellulose-nano magnetite $Fe3O4$ nanocomposites	Synthesis of methyl esters from oleic acid	Conversion yield of 89.21\%	Anggraini (2019)
Carboxylated cellulose doped with Sn (II) ions	Transesterification and hydrolysis of soybean oil and esterification of fatty acids	Conversion yield $= 55\%, 93\%$ and 95% respectively	Santos et al. (2016)
Cellulose matrix embedded copper decorated magnetic bionanocomposite	Synthesis of dihydropyridines and polyhydroquinolines	Conversion yield $=$ More than 85%	Maleki et al. (2019)
Cellulose@hematite-zirconia nanocomposite	Production of biodiesel through the esterification of biomass-derived materials	Conversion yield $= 92.5\%$	Helmiyati et al. (2021)
Novel magnetic biobased heteropolyacid prepared via in-situ reaction	Transesterification of tree-born oil	Conversion yield $= 96.6\%$	Bao et al. (2021)

Table 4 Nanostructured cellulose-based materials used as a catalyst in synthetic reactions

Presently, biopolymer-based flocculants which are mostly being used and are gaining attention include chitosan, cellulose, tannin and alginate. Bioflocculant is not only beneficial for their biodegradability, but these materials have dimensions in the nanorange and also possess a high surface specific area (Haver and Nayar [2017\)](#page-26-29). Natural polymer-based flocculants can be categorized into two groups: (i) grafted natural polymers to develop semi-natural flocculants, (ii) modified natural polymers to design natural flocculants (Zhu et al. [2015;](#page-27-7) Das et al. [2013\)](#page-23-25). Nanocelluloses are still not much frequently used as flocculants. As discussed in the previous sections, researchers majorly use these materials either as an adsorbent or as a photocatalyst. The main issue of using CNCs as flocculants is their tendency of agglomeration because of the networks developing by the formation of hydrogen bonds between –OH groups on the surface of CNCs (Yu et al. [2016b\)](#page-27-8). The adjustment in the surface –OH group changes the properties of CNC flocculants and prevents them from agglomerating. Nanocellulose obtained from the processing of pulp acquires a negative charge on its surface. As a result, anionic nanocellulose does not interact strongly with negatively charged inorganic minerals. Moreover, anionic flocculants are poorly soluble in an acidic medium and, therefore, cannot be used in these conditions (Kono [2017\)](#page-24-20). The chemical pre-treatment of CNCs applying different methods such as carboxymethylation (Liimatainen et al. [2014\)](#page-24-21), citric/hydrochloric acid hydrolysis (Habibi et al. [2010\)](#page-23-26), periodate-chlorite oxidation (Yu et al. [2016b;](#page-27-8) Liimatainen et al. [2014\)](#page-24-21), TEMPO-mediated (Suopajärvi et al. [2017\)](#page-26-27) etc. is done for increasing the density of negative charge or furnishing nanocellulose with cationic

charges. Cationic counterparts of nanocellulose assist the removal of anionic dyes through electrostatic interaction of oppositely charged particles.

Currently, researchers are working on the functionalization of nanocellulose by instigating anionic, cationic or hydrophobic functional groups onto the surface of CNCs through the mechanism of charge neutralization. The presence of -OH groups on the surface of cellulose facilitates the insertion of desired functionality on their surface and thereby forms an effective flocculant (Azizi et al. [2013\)](#page-22-20). The application of flocculants for wastewater treatment is presented in detail in Table [5.](#page-18-0) Suopajarvi and co-workers reported nano cellulosic materials functionalized with anionic and cationic di-aldehyde as water chemicals in wastewater flocculation (Liu et al. [2012,](#page-24-22) [2015;](#page-24-15) Ma et al. [2017b\)](#page-25-18). Productive results were obtained, which proved the potential of flocculants for wastewater treatment. The remodeling of CNCs was done by periodate oxidation through which the aldehyde group was introduced by oxidation of surface –OH groups and modification in the carbon network of glucopyranose ring (Wang et al. [2014b\)](#page-27-9). Liimatainen et al. examined the application of both anionic and cationic di-aldehyde cellulose generated via aqueous periodate oxidation (Liimatainen et al. [2011,](#page-24-23) [2012\)](#page-24-24). The study revealed that the flocculation performance of kaolin was better using anionic cellulose nanoparticles as compared to cationic ones.

CNC based flocculant	Pollutant	Flocculant performance	References
CNCs modified with $1-(3-aminopropyl)$ - imidazole (APIm)	Microalgae (Chlorella <i>vulgaris</i>)	Efficiency $= 100\%$, Maximum adsorption capacity = $43.4 g$ algae/g	Ge et al. (2016)
Anionic dicarboxylic acid nanocellulose	Municipal wastewater	Turbidity reduction $=$ 80% ^a , COD removal = 60%	Kimura and Itoh (1996)
Dicarboxylic acid CNFs	Municipal wastewater solid particles	Turbidity reduction $=$ 40–80%, ${}^{\rm a}$ COD removal = $40-60\%$	Suopajarvi et al. (2013)
Rod-shaped cellulose nanocrystals	Flocculation and phase separation of bacteria	Aggregation percentage $=100\%$	Vanderfleet et al. (2018)
Ouaternized CNFs	Reactive orange 16	maximum adsorption $capacity =$ 0.477 mmol/g for reactive orange 16	Haver and Nayar 2017)

Table 5 Flocculation of water contaminants using water-based flocculants

^a COD–Chemical Oxygen Demand

5.6 Nanocellulose-Based Membranes

This section elaborates on the fabrication of nanocellulosic membranes and their role in environment remediation. Nanocellulosic membranes are constituted of both cellulose nanofibers and cellulose nanocrystals. The fabricated membranes are employed not only for wastewater treatment (Cruz-Tato et al. [2017\)](#page-23-28) but also for gas separation (Ansaloni et al. [2017\)](#page-22-21), dye removal (Karim et al. [2014\)](#page-24-25), removal of heavy metal ions through ultrafiltration (Karim et al. [2016a,](#page-24-26) [b\)](#page-24-27), adsorption (Karim et al. [2017\)](#page-24-28) and ion exchange process (Chitpong and Husson [2017\)](#page-23-29). In this regard, Cruz-Tato et al. [\(2017\)](#page-23-28) reported metal-based nanocellulose composites for developing a membrane capable of supporting forward osmosis for the removal of pollutants from water bodies. The change of nanocellulose was done to attach the functionalities of amino silane before the coating of platinum and silver nanoparticles. The fabricated membrane was more efficient as compared to HTI-CTA membrane (Cellulose acetate membrane from Hydration Innovation Technologies) in terms of water flux but shows the contrasting result for total organic carbon rejection. In contrast to this, a highly productive and high capacity cation exchange membrane was developed by Chitpong and Husson [\(2017\)](#page-23-29) for the elimination of trace Cd (II) ions discharged in water bodies.

The removal of contaminants via membranes is greatly influenced by the pore size. For this reason, membranes are developed in such a way that the size of their pores excludes the contaminants allowing only purified water to pass through. Several researchers have employed nanocellulosic papers derived from plants and algae for the removal of water pollutants via size based exclusion. Bismarck and co-workers prepared nanopaper from plant sources (Mautner et al. [2014,](#page-25-22) [2015\)](#page-25-23). In their work, two different types of nanopapers were prepared using CNF-0 having a fibril diameter of 5–30 nm and CNF-K of fibril diameter of 50 to 100 nm. It was observed that the permeability of nanopapers prepared using CNF-K could be regulated over a wider range owing to larger pore dimensions.

The chemical functionality of membranes also has an influence on the membrane filtration process. The membrane can be made selectively permeable for chemically mediated interactions by regulating the charge present on its surface. Mathew and co-workers used cellulose nanomembrane for charge mediated adsorption of water contaminants (Karim et al. [2014,](#page-24-25) [2016b,](#page-24-27) [c\)](#page-24-29).

Apart from using these techniques, individual simultaneous application of these techniques has also been reported by researchers. Researchers from Hsiao and Chu's group were successful in removing several types of water pollutants simultaneously using nanocellulose membrane by applying both the principles of size based exclusion and charge mediated adsorption in a single process (Wang et al. [2013;](#page-27-10) Ma et al. [2011,](#page-24-30) [2012\)](#page-24-31). Several other membranes are listed in Table [6.](#page-20-0)

Cellulose type	Membrane application	Membrane performance	References
Chitosan based cellulose nanocrystal	Removal of dye	Removal percent: 69-98%	Karim et al. (2014)
TFNC membrane infused with carboxylated CNCs	Removal of bacteria $(E. coli, B. diminuta)$, virus (Bacteriophage MS2) and organic dye (crystal violet)	Average pore size $=$ $0.22 \mu m$; Porosity = 80% ; Pressure drop = 3.0 kPa; Permeability $=$ 59 Lm ⁻² h ⁻¹ kPa ⁻¹ : $LRV = 6$ for bacteria and 2 for MS2 virus: maximum adsorption capacity for crystal violet $=$ 0.166 mmol/g	Ma et al. (2012)
PAN composite membrane blended with carboxylated CNCs	Removal of silica particles (7–40 nm), oil water emulsions	Pore size $=$ 5–60 nm; Rejection ratio $=$ > 99%	Cao et al. (2013)
Nanopaper made from pristine CNFs	Filtration of swine flu influenza virus $(80-120 \text{ nm})$ and polystyrene latex beads of size(30, 100, 500 nm)	Average pore size $=$ 19 nm; Porosity = 35% ; $SSA = 88 \text{ m}^2/\text{g}$; Flux = $50 \mu Lh^{-1}$ cm ⁻²	Metreveli et al. (2014b)
Metalized nanocellulose composites-thin film composite membrane (MNC-TFC)	Wastewater treatment (to remove urea)	water flux (LMH/bar): 1. urea: 7.4; wastewater: 11.8 2. urea: 5.4; wastewater: 11.5 (both membranes performed better than a commercial HTI-CTA membrane)	Cruz-Tato et al. (2017)

Table 6 Application of nanocellulose-based membrane in membrane technology for wastewater treatment

6 Limitations and Future Prospects

Although nanocellulose has proven to be successful in addressing the concerns related to wastewater treatment, there are still several challenges and disadvantages associated with it which needs to be resolved. The commercialization of nanomaterials can be promoted only if these materials are prepared on a large scale at a low cost and generate valuable end products (Carpenter et al. [2015\)](#page-22-3). Most of the works reported on the preparation of nanocellulose are at a laboratory scale. The major challenges with selecting the treatment method for fabricating nanocelluloses are the larger scale of production, lower cost and environment feasibility. In order to upscale the production, several techniques related to mechanical disintegration are applied by researchers. But the higher energy consumption is the drawback associated with these techniques. Although a lot of effort is being invested in discovering other mechanical techniques,

most of them are still at a primitive stage and are not applicable for upscaling. In contradiction to this, Arvidsson et al. [\(2015\)](#page-22-23) reported that chemical pre-treatment is not necessarily less energy consuming for generating nanocellulose. In this study, a chemical treatment known as carboxymethylation was carried out, which exhibited that a higher energy consumption occurs in chemical pre-treatment as compared to applying only homogenization without any pre-treatment because of the high intake of chemicals such as methanol, ethanol, and isopropanol for the production of nanocellulose (Arvidsson et al. [2015\)](#page-22-23). In this case, the consumption of electricity is low, but the presence of acidic and solvent waste results in a higher environmental impact (Carpenter et al. [2015\)](#page-22-3). The second challenge is the production of bacterial and nanofibrillated cellulose at a large scale. The reason for this uncertainty is the dependence of production on the build up of nanofibers from low molecular weight sugars by bacteria and dissolved cellulose by elecrospinning, respectively (Nechyporchuk et al. [2016\)](#page-25-8). The strategies of production need strict vigilation to formulate an eco-friendly and economically viable method for the production of nanocellulose. Furthermore, the undefined toxicology of modified nanocellulose materials creates economic obstacles for its application in wastewater treatment. In general, the reduction in the size of nanoparticles increases the risk of their inhalation to cells, lymph and blood circulation and damage the potentially sensitive target area (Oberdörster et al. [2005\)](#page-25-25). Although most of the research work shows that nanocellulose-based materials are non toxic (or slightly toxic) in nature, researchers have also reported that at higher concentrations, these materials have a negative impact on cell viability and proliferation (Alexandrescu et al. [2013\)](#page-22-24). Moreover, prolonged inhalation of these nanofibers may have cytotoxic and inflammatory effects on human lung cells and also on the lung cells of mice, as reported by Yanamala et al. [\(2014\)](#page-27-11).

7 Conclusion

The recognition of nanocellulosic materials may be considered a major milestone in the field of Materials Science and Engineering. A lot of attention has been grabbed by these nanocellulosic materials owing to their application in several fields such as bio-based food packaging, tissue regeneration, optoelectronics and environmental remediation, to name a few. Cellulosic nanomaterials possess properties such as large surface area, facile functionality, and eco-friendly nature (renewability and low toxicity), which makes them suitable for application in water and wastewater treatment. As discussed in the chapter, cellulosic nanomaterials effectively remove the pollutants present in water bodies through different methods such as adsorption, flocculation, membrane formation, and catalyst carrier. The challenges associated lies with the percentage yield and energy consumed during the stage of preparation and toxicity of the product obtained at the end. Both these issues were simultaneously sorted by applying an integrated method for the preparation of nanocellulose. The alteration in the structure of these nanocelluloses improved its functionality, making it even more efficient for the removal of pollutants. In spite of several advantages, there is still a possibility to develop further improved treatment schemes for effective extraction of nanocellulose and its functionality as agents to be applied not only for wastewater treatment but also for other environmental remediation with reference to economic feasibility and its performance for wastewater treatment.

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