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Cellulose-Based Materials: A Comprehensive Review for Sustainable Water Treatment Solutions

Deeksha Ranjan[1], Richa Choudhary[2]*
[1]Department of Applied Sciences and Humanities, Faculty of Engineering and Technology,
Rama University, Mandhana, Kanpur, U.P.-209217
Email: deeksha.rs.apc@itbhu.ac.in
[2]*Department of Bio-Sciences, Division of Life Sciences, School of Basic and Applied Science,
Galgotias University, Greater NOIDA, U.P.-203201.
Email: richa.choudhary@galgotiasuniversity.edu.in

Abstract

Cellulose, a ubiquitous biopolymer derived from plant cell walls, finds extensive application in water treatment processes due to its remarkable properties. This abstract explores the diverse uses and benefits of cellulose in water treatment applications. Cellulose-based materials serve as effective adsorbents for removing various contaminants from water, including heavy metals, organic pollutants, and dyes, owing to their high surface area, porosity, and functional groups. Moreover, cellulose derivatives such as carboxymethyl cellulose and cellulose acetate enhance water treatment efficiency through membrane filtration processes, facilitating the separation of impurities from water streams. The eco-friendly nature of cellulose-based materials aligns with the growing demand for sustainable water treatment solutions, offering a renewable alternative to conventional synthetic polymers. Furthermore, the facile modification and functionalization of cellulose enable tailored designs for specific water treatment needs, enhancing performance and selectivity. Overall, cellulose-based materials exhibit immense potential in addressing water quality challenges, contributing to the development of efficient, cost-effective, and environmentally friendly water treatment technologies.

1. Cellulose based techniques for wastewater treatment:

Cellulose, one of the most found natural polymers on earth, consists of repeated units of β-D-glucopyranose linked through β-1,4-glycosidic linkage and derived from various wood sources. In its natural form cellulose has lesser applications. The functional groups present on cellulose surface can be modified with the help of physical and chemical methods and the derivatives are then employed for various applications including wastewater treatment. It may be modified by directly introducing inorganic nanoparticles as metal oxides, chelating agents or metal binders or by grafting of selected monomers into the cellulosic backbone as biopolymer chitosan (1-3). With modification and fabrication, cellulose can be employed for many commercial applications and products as membranes etc. Cellulose based nanomaterials can be prepared via various physical, chemical, biological and mechanical methods. Nanoscale derivatives include cellulose nanocrystals, cellulose nanofibrils, cellulose nanofibrils (CNFs), and TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl radical)-mediated oxidized cellulose nanofibrils (TOCNF) (4, 5). The below section shows the possibility of cellulose and cellulose based materials in various wastewater treatment technologies.

1.1 Cellulose and its derivatives in precipitation:

Precipitation of water contaminants, such as metal ions and anionic species, is one of the oldest methods in water treatment. Precipitation of contaminants is carried out by adding various chemical precipitating reagents including cellulose. Cellulose has been reported to be used for the co-precipitation of various metal ions as Cd(II) (5). The precipitated pollutants are then removed via decantation or filtration. The filtration is carried out by filter paper or membrane. Commercial filter papers like Whatman® filter paper are made up of cellulose.

1.2 Cellulose and its derivatives for adsorption:

The main feature of cellulose is the presence of hydroxyl ions on its surface, which show affinity towards adsorption of heavy metals ions and organic ions. Raw agricultural residues which contain cellulose have low load capacity as well as selectivity as the -OH groups are involved in intermolecular hydrogen bonding (6). These hydrogen bonds can be destroyed by chemical modification of cellulose decreasing their crystalline structure and making the surface binding sites more efficient as -OH groups become more active to bind with other functional groups, finally enhancing the adsorptive property of cellulose. Variously modified cellulose, grafted cellulose, cellulose beads and composites, nanocellulose, and cellulosic hydrogels have been employed for adsorption of organic and inorganic water pollutants (6) as shown in below table:

Table 3: Cellulose and derivatives used in adsorption of various water pollutants

Adsorbent	Pollutant	Maximum adsorption capacity	Year	Reference
Neutral and variously modified cellulose nanofibrils	Multiple active pharmaceutical ingredients	1.3 mg/g for MTF to 11.8 mg/g for DCF by unmodified LNPs	2022	9
Chemically modified cellulose fibers (PEI/CE)	Acid fuchsin (AF), Cu(II), perylene tetracarboxylate (PTC), and Zn(II) ions	AF - 562 mg/g Cu(II) - 552 mg/g PTC - 216 mg/g Zn(II) - 157 mg/g	2022	10
Sulfated Cellulose nanocrystals	Janus Green (JG) dye	77 mg g^{-1}	2022	11
Spherical cellulose-based adsorbent (2-AMP resin) having 2-aminomethyl-pyridine	Ag(I)	-	2020	12
Composite nanofiber membrane of Deacetylated cellulose acetate and polydopamine	methylene blue (MB)	88.2 mg/g	2020	13

Cellulose isolated from straw was modulated using N(4)-morpholinothiosemicarbazide	Ni(II)	90 mg g–1	2020	14
Thiosemicarbazide-modified cellulose	Cu(II)	106.3829 mg g–1	2020	15
Sodium Alginate/Hydroxypropyl Cellulose Beads	Pb (II)	47.72 mg/g	2020	16
macrocyclic pyridone pentamer (MCPP) with succinic acid anhydride modified microcrystalline cellulose (SA-MCC)	Ba(II), Pb(II), Cd(II), Mn(II), Cu(II), Co(II), Ni(II), Cr(III)	75% of Ba2+ and others less than 18%	2019	17
Thiosemicarbazide-modified cellulose	(Hg)II	98.5% at a pH 5.0	2019	18
Grafting amino-terminated hyperbranched polymer (NH2-HBP) and beta-cyclodextrin (β-CD) onto cotton fibers	Congo red (CR) and methylene blue (MB)	350.8 mg/g for CR and 102.7 mg/g for MB	2019	19

Sulfonated cellulose	Cu2+	8.2 mg-Cu2+/g	2017	20
Cellulose nanocrystal-alginate hydrogel beads	Continuous flow adsorption of methylene blue	255.5 mg/g	2015	21
Cellulose nanocrystals and alginate (CNC–ALG hydrogel beads	methylene blue (MB)	256.41 mg/g	2015	22
Carboxylated cellulose nanofibrils (CCNFs)	Pb(II)	171.0mg/g	2014	23
Hydrogels based on cellulose-graft-poly (acrylic acid) copolymers (C-g-AA)	Cu(II) and Ni(II)	182 and 200 mg/g for Cu(II) and Ni(II), respectively	2016	24
Cellulose formed with vinyl monomer glycidyl methacrylate by using ceric ammonium nitrate and functionalized with thiosemicarbazide	Hg(II), Cd(II), and acid fuchsin (AF)	-	2013	13

Branched poly (ethylene imine) (PEI)-modified cellulose-based adsorbent (Cell-g-PGMA-PEI)	Cu(II)	102 mg g−1	2012	25
Cellulose wood pulp grafted with vinyl monomer glycidyl methacrylate (GMA) and functionalised with imidazole (cellulose-g-GMA-imidazole)	Pb(II), Ni(II)	72 mg g-1 of Pb(II) and 45 mg g-1 of Ni(II)	2006	26
Carboxymethylcellulose (CMC)	Cu2+	-	2004	27

Differently modified, gafted cellulose hydrogels, beads, and resins have been employed for the adsorption of water pollutants (28-32). O’Connell et al., 2008 (32) have listed a number of adsorbents prepared from modulation of cellulose for remediation of heavy metal ions. Hokkanen et al., 2016 (29) have reviewed various methods utilized to modulate cellulose based adsorbents to enhance their adsorptive property. A review published in 2021, by Akter et al. (30), concisely reviews the cellulose based hydrogels for wastewater treatment. The article reviews many heavy metal ions and organic dyes which have been adsorbed on the surface of cellulose based hydrogels. These various kinds of hydrogels based on cellulose have been reported to be superadsorbents and very durable, non-toxic, biodegradable and almost complete adsorption of heavy metal ions, dyes etc. from water as well as wastewater (33).

1.3 Cellulose and its derivatives for coagulation/ flocculation:

The main issue in addition of chemical coagulants/ flocculants as aluminium chloride and ferric chloride is the production of secondary pollutants. This can be resolved by employing biopolymers like cellulose as coagulant, which is eco-friendly and requires comparatively lesser steps. Cellulose based coagulants and flocculants are obtained by either by physically combining it with other material or by chemically modifying them or both (34,35). In research conducted by Mohamed Noor et al. in 2018 (36) cellulose was hybridized with a magnetite powder to form a flocculant to treat effluents from palm oil mill. Cellulose backbone has been modified with Ag2O and TiO2 and has been used for photodegradation (35). In a study carried out in 2010 (37) an anionic sodium carboxymethylated cellulose was formed from date palm rachis. It was combined with aluminium sulphate coagulant and was used as flocculant to remove turbidity while treatment of drinking water. Another study reports the use of anionized nanocellulose as flocculant combined with ferric sulphate coagulant in the treatment of municipal wastewater (38). There are reports of using sawdust derived cellulose nanocrystals as coagulant for Ni(II) and Cd(II) from water, showing adsorption power of 956.6 mg/g for Ni2+ and 2,207 mg/g for Cd2+ (39). It is also reported that modification with hexadecyltrimethylammonium bromide (HDTMA-Br) showed better performance in comparison to the commercially available coagulant R_2T_2 (40). A review published in 2021, by Fauzani et al. (41), mentions cellulose in natural flocculant applications. Another review article by Barrero-Fernández et al., in 2021 (42) does a bibliometric analysis on the application of cellulose-based materials used for flocculation.

1.4 Cellulose and its derivatives for membrane filtration:

In general, the physicochemical properties of membranes can be enhanced by application of chemical nanomaterials, which have been found to be toxic in nature. This has led to the use of low-cost, eco-friendly, and renewable cellulose or nanocellulose. Membranes for commercial purposes are fabricated by the use of cellulose like, polysulfone, polyethersulfone, polyvinylidene fluoride, polypropylene (43, 44). In recent years nanocellulose, like cellulose nanocrystals, nanofibrils, bacterial nanocellulose have also been in application in development of different types of membranes, such as forward osmosis, membrane distillation, reverse osmosis, nanofiltration. Membranes based on nanocellulose have been found to be effective in desalination and thus commercial water/ wastewater treatment. Chemical modification of cellulose/ nanocellulose surface has also been found to be enhancing the surface properties and thus its affinity and reactivity towards effective removal of contaminants from water (Table 4) (45-47).

Table 4: Cellulose and derivatives used in membrane filtration for various water pollutants

Membrane	Contaminants	Rejection rate/ Remarks	Year	Reference
Application of Cellulose Nanocrystals in Nanofiltration				
Nanocomposite membrane containing polydopamine-modified cellulose nanocrystals	Congo red (CR)/NaCl solution	Congo red rejection - 99.91% Salt permeation - 99.33% Mixture congo-red/NaCl selectivity - 98%	2020	48
Nanofiltration membrane from bamboo cellulose	NaCl	36.11 %	2021	49
Polyamide nanofiltration membrane based on Cellulose	MgSO4	Enhanced about 9.3%	2020	50
Thin film nanocomposite nanofiltration membrane incorporated with cellulose nanocrystal/silver nanocomposites into a polyamide layer	Na2SO4	99.1% for Na2SO4. Also 99.4% reduction of *Escherichia coli* viability	2020	51

Thin film composite nanofiltration membranes modified by cellulose nanocrystal sandwiched layer and a polydopamine layer	Congo red, Rose Bengal, sodium lignosulfonate and alkaline lignin	Excellent	2020	52
2,2,6,6-tetramethylpiperidine-1-oxyl radical (TEMPO)-oxidized cellulose nanofibers (CNFs) incorporated into the polyamide layer of thin film composite (TFC) reverse osmosis (RO) membrane	NaCl	96.2%	2019	53
Cellulose nanocrystals (CNCs) incorporated into polyamide (PA) layer to prepare thin film composite (TFC) nanofiltration membranes (CNC-TFC-Ms)	Na2SO4 and MgSO4	98.0% and 97.5% for Na2SO4 and MgSO4, respectively	2019	54
Cellulose nanocrystals incorporated polyamide layer of thin film composite membrane	Na2SO4 and MgSO4	98.7% and 98.8% respectively	2018	55

Triple-layered composite nanofiltration membranes by interfacial polymerization of diamine and acyl chloride on cellulose nanocrystal interlayer supported by microporous substrate	Na2SO4, MgSO4, MgCl2, CaCl2, NaCl	Na2SO4 (97.7%) > MgSO4 (86%) > MgCl2 (15.5%) > CaCl2 (11%) > NaCl (6.5%)	2017	56
Application of Cellulose Nanocrystals in Reverse Osmosis				
Carboxylated cellulose nanocrystal (CNCCR) incorporated active layer (AL-CNCCR) and supporting layer (SL-CNCCR) of TFC membrane	Na2SO4, MgSO4, NaCl	AL-CNCCR rejection ratio increased to approximately 105.2%, and SL-CNCCR membranes showed 107.5% rejection ratio for sulphate ion. Also for sodium chloride; the increased value for chloride ion rejection of SL-CNCCR membrane (134.4%) and with AL-CNCCR membrane (108.7%)	2021	57
Sawdust-derived cellulose nanocrystals embedded in polyamide	sodium chloride, and calcium chloride	sodium chloride - 98.3 ± 0.8% calcium chloride - 97.1 ± 0.5%	2020	58
Cellulose nanocrystal membrane grafted with Poly(acryloyl hydrazide)	-	better water permeance, salt rejection, and organic fouling resistance, improved boron rejection	2019	59

Reverse Osmosis nanocomposite membrane incorporated with 2,2,6,6-Tetramethylpiperidine-1-oxyl oxidized cellulose nanocrystals	Salt	98.98 ± 0.41%	2019	60
Nanocomposite membrane incorporated with modified cellulose nanocrystals	Synthetic brackish water	97.8%. Also, good fouling resistance properties for tested Bovine Serum Albumin	2018	61
Application of Cellulose Nanocrystals in Pervapouration (PV)				
1.Nanocomposite membranes based on poly(styrene)-block-poly(butadiene)-block-poly(styrene) (SBS) matrix and CNCs (SBS/CNC membranes) 2. decorated the CNCs with hydrophobic oleic acid moieties (OLA-CNCs) (SBS/OLA-CNC membranes)	Ethanol	1. SBS/CNC membranes exhibit consistent reduction towards ethanol selectivity 2. SBS/OLA-CNC increased ethanol permeability	2021	62
CTA/CNCs membrane	NaCl hypersaline solution	99.8%	2021	63

Functionalized cellulose nanocrystal incorporated TFN's polyamide thin layer	-	Good resistance for protein fouling and reduced water flux drop	2020	64
Cellulose triacetate/cellulose nanocrystals nanocomposite DMSO based pervaporation membranes	NaCl	99.9%	2020	65
Cellulose triacetate/cellulose nanocrystals (CTA/CNCs) nanocomposite PV membrane.	NaCl	99.9%	2019	66
Application of Cellulose Nanofibers in Nanofiltration				
Nanofiltration Membranes on blended metal ion support	Dye and salt	Dye rejections for Congo red - 97.7%, Methyl blue - 97.1%, Eriochrome Black T - 95.0% Salt penetration rate for Na_2SO_4 - 93.8%, $MgSO_4$ - 95.1%, NaCl - 97.4%,$MgCl_2$ - 98.1%	2022	67
Nanofiltration membrane composited with sulfated cellulose nanofibril	Na2SO4	Above 98%	2022	67

Oxygen Plasma Treated composite membrane of Reduced Graphene Oxide/Cellulose Nanofiber	Acid Fuchsin, Rose Bengal, Brilliant Blue	Above 90%	2021	68
Application of Cellulose Nanofibers in Ultrafiltration				
High-lignin unbleached neutral sulfite’s cellulose nanofibers for ultrafiltration	-	water flux efficiency was 96% higher	2020	69
Surface modification via interfacial polymerization of ultrafine cellulose nanofiber (UCN) membranes	MgCl2, MgSO4, NaCl , Na2SO4	MgCl2—89.7%, MgSO4—65.3%, NaCl—43.6%, and Na2SO4 (39.1%)	2017	70
Application of Cellulose Nanofibers in Reverse Osmosis				
Reverse osmosis membranes of aromatic polyamide (PA) reinforced with a crystalline cellulose nanofiber (CNF)	Chloride ions	higher hydrophilicity and higher water permeability, enhanced antifouling performance and improved chlorine resistance	2020	71
Composite reverse osmosis membrane of polyamide layer incorporated with 2,2,6,6-tetramethylpiperidine-1-oxyl	NaCl	96.2%	2019	72

radical (TEMPO)-oxidized cellulose nanofibers				
Oxidized cellulose nanofibers membrane modified with Cu-terpyridine	Cr(VI	80%	2018	73
Cellulose Acetate/Cellulose Nanofiber Membranes	whey, strawberry juice and raspberry juice	Reduced turbidity and solid content	2017	74
Application of Bacterial Nano Cellulose (BNC) in Membrane distillation				
Polydopamine particles and bacterial nanocellulose advanced membrane	salt	>99.9%	2021	75
Unsupported bacterial nanocellulose aerogel membranes	-	higher intrinsic membrane permeability and thermal efficiency	2016	76
Application of Bacterial Nano Cellulose (BNC) in Ultrafiltration				

Membrane composed of polydopamine (PDA) particles and bacterial nanocellulose (BNC)	Pb, Cd, Rhodamine 6G (R6G), methylene blue (MB), and methyl orange (MO)	easy to fabricate, highly scalable, chemically stable, mechanically robust, and reusable	2019	77
Ultrafiltration membrane of reduced graphene oxide and bacterial nanocellulose	-	two-fold increment of water flux	2019	78
Membrane based on bacterial nanocellulose (BNC) loaded with graphene oxide (GO) and palladium (Pd) nanoparticles	Methyl orange, Multiple contaminants (4-nitrophenol, methylene blue, and rhodamine 6G)	highly efficient methylene orange (MO) degradation during filtration (up to 99.3%	2018	79

Conclusion

In conclusion, this review highlights the significant role of cellulose-based materials in advancing sustainable water treatment solutions. Through an exploration of various applications and properties, it is evident that cellulose offers versatile benefits for addressing water quality challenges. From its ability to adsorb contaminants to its effectiveness in membrane filtration processes, cellulose demonstrates promise as a renewable and eco-friendly alternative to traditional materials. The review underscores the importance of cellulose derivatives, such as carboxymethyl cellulose and cellulose acetate, in enhancing water treatment efficiency and selectivity. Moreover, the facile modification and functionalization of cellulose enable tailored designs to meet specific water treatment needs. As society increasingly prioritizes environmental sustainability, the use of cellulose in water treatment aligns with these objectives, offering a renewable and biodegradable solution. Moving forward, further research and development in cellulose-based materials hold potential for innovation in water treatment technologies, paving the way for more efficient, cost-effective, and environmentally friendly solutions to ensure access to clean water for all.

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