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REVIEW

Remediation of heavy metals and dyes from wastewater using cellulose-based adsorbents

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Abstract

Heavy metals and dyes are major pollutants that pose potential threat to the health of humans and ecosystems. Various technologies are available to remediate such pollution, but these processes are costly, have high energy requirements and generate toxic sludges and wastes that need to be carefully disposed. There is therefore a need for methods that are more efficient, cost effective and environment friendly for water purification. Adsorption is regarded as a green, clean and versatile method for wastewater treatment. In particular, biodegradable and non-toxic materials such as cellulose-based materials are of interest for water purification. Moreover, the surface of cellulose contains many hydroxyl groups that facilitate the incorporation of chemical moieties, thereby improving pollutant adsorption. Here, we review the most relevant applications of cellulose-based materials for wastewater treatment. A major point is that reducing cellulosic dimension to nanometric levels highly improves adsorption of heavy metals and dyes from wastewaters. Nanocellulose and functionalized nanocellulose are thus promising for wastewater treatment.

Keywords Heavy metal · Dye · Adsorption · Green adsorbents · Cellulose · Nanocellulose

Introduction

Issues related to the quality of water are one of the major problems faced by humanity in the twenty-first century. The quality of our water resources is deteriorating day by day due to various anthropogenic activities, increasing industrialization and unplanned urbanization. Among the various organic and inorganic pollutants encountered in wastewater, toxic heavy metals and dyes are among the major pollutants that pollute the aquatic environment. Wastewater effluents containing dyes and heavy metals cause potential hazard to the environment and human health. Recently, numerous approaches such as ion exchange, membrane separation, electrochemical treatment and adsorption (Barakat 2011) have been studied for the development of effective technologies to decrease the amount of wastewater produced and to improve the quality of the treated effluent. However, most of them require substantial financial input and thus their use is restricted. Among all the treatment processes mentioned, adsorption is found to be effective, simple and relatively lower operation cost of pollutant removal (Meshko et al. 2001). Different types of materials such as activated carbon, zeolites, carbon nanotubes and molecularly imprinted polymers have been developed as adsorbents. In recent years, development of green adsorbents received widespread attention as they are valued for their renewability, low cost and non-toxicity (Chang and Juang 2004). Green adsorbents are manufactured in a more energy conservative way, pose few health problems and are recyclable. Cellulosic adsorbents have the proficiency to meet almost all the requirement for being green. Cellulose is the most abundant, natural biopolymer which is renewable, biodegradable and non-toxic. The primary occurrence of cellulose is the existing lignocellulosic material. One of the promising applications of lignocellulosic material is as an adsorbent for water

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purification or wastewater treatment due to its wide availability, renewability, sustainability and the possibility of surface modification. Several researches have been devoted to review the removal of organic and inorganic pollutants using lignocellulosic adsorbents (Abdolali et al. 2014). However, untreated lignocellulosic biomass is generally not functional and the adsorption capacity varies depending on the biomass source. When the size is minimized to the nanoscale, the high specific surface area of the polysaccharide adsorbents contributes to enhancing the adsorption capacity. This led to the emergence of nanocellulose as a new generation of bio-based adsorbents with potential applications in wastewater treatment. These nanomaterials have been extensively explored by researchers as an adsorbent for removal of various kinds of hazardous pollutants and the studies indicate that these materials possess high adsorption capacity, are environmental friendly and inexpensive (Lam et al. 2012). Cellulose-based materials are more attractive for water purification when it makes structural modifications to improve their existing properties or adding new potentialities to this material (Silva Filho et al. 2013). Benefiting the presence of abundant hydroxyl groups on the surface of micro- or nanocellulose offers a unique platform for significant surface modification to graft a myriad of functional groups or molecules onto the cellulosic structure thereby immobilizing pollutants. Here, we review the recent progresses related to the application of cellulose-based materials and their modified forms as an adsorbent for the removal of toxic heavy metals and dyes from wastewater. Herein the adsorption efficacies of various green adsorbents, cellulose-based green adsorbents, modified cellulose-based adsorbents and modified nano/micro-based adsorbents have been discussed. This article is an abridged version of the chapter published by Varghese et al. (2018) in the series Environmental Chemistry for a Sustainable World (https://www.springer.com/series/11480).

Heavy metals

The term heavy metal refers to any metallic element that has a density more than 5 g per cubic centimeter and is toxic or poisonous even at low concentration. These include lead (Pb), cadmium (Cd), zinc (Zn), mercury (Hg), arsenic (As), silver (Ag), chromium (Cr), copper (Cu), iron (Fe) and the platinum group elements. Most of the metals are non-biodegradable, highly toxic and carcinogenic in nature (Barakat 2011). Heavy metals cause serious health effects, including reduced growth and development, cancer, organ damage, nervous system damage, and in extreme cases, death. At higher doses, heavy metals can cause irreversible brain damage. Therefore, it is necessary to treat metal-contaminated wastewater before its discharge into the environment. Table 1 (Abdel-Raouf and Abdul-Raheim 2017) lists those heavy metals that are relevant in the environmental context, their source and toxicity effect.

Dyes

Dyes are complex organic compounds which are purged from various industrial sources such as textile, cosmetic, paper, leather, rubber and printing industries to color their products. To meet industrial demand, it is estimated that 1.6 million tons of dyes are produced annually and 10–15% of this volume is discarded as wastewater. As a result, dyes are major water pollutants. Excessive exposure to dye causes skin irritation, respiratory problems and some dyes even increase the risk of cancer in humans (Rai et al. 2005). Thus, it is of utmost importance to remove dyes from wastewater effectively to ensure safe discharge of treated liquid effluent into watercourses.

Table 1 Source, route of entry and toxicity effect of some heavy metals

Heavy metal	Source	Route of entry	Toxicity effect
Arsenic	Pesticides, fungicides, metal smelters	Inhalation and ingestion	Irritation of respiratory system, liver and kidney damage, loss of appetite, nausea and vomiting
Cadmium	Welding, electroplating, pesticide fertilizer, Cd–Ni batteries	Inhalation and ingestion	Lung, liver and kidney damage; irritation of respiratory system
Chromium	Paints, electroplating and metallurgy	Inhalation, ingestion, and absorption through skin	Lung damage and irritation of respiratory system
Mercury	Pesticides, batteries, paper industry	Inhalation, ingestion and absorption through skin	Irritation of respiratory system; lung, liver kidney damage, and loss of hearing and muscle coordination
Lead	Paint, pesticide, smoking, automobile emission, mining	Inhalation and ingestion	Lung and liver damage; loss of appetite, nausea
Nickel	Electrochemical industries	Inhalation	Lung, liver and kidney damage

Methods for pollutant removal

In response to the rising demands of clean and safe water, many different technologies are available for treating the pollutant-laden wastewater. Some of the widely used treatment technologies include biological treatments (McMullan et al. 2001), membrane process (Barakat 2011), chemical and electrochemical technology (Ku and Jung 2001), reverse osmosis (Sonune and Ghate 2004), ion exchange (Maranon et al. 1999), electrodialysis, electrolysis and adsorption procedures (Barakat 2011). Table 2 collects some of the common technologies that have been adopted by researchers for the removal of heavy metals and dyes from wastewater along with their advantages and disadvantages. Among the various decontamination techniques, adsorption process is regarded more prospective for water treatment due to its ease of operation, convenience and simplicity of design (Faust and Aly 1981).

Adsorption

The general mechanism of adsorption involves the transfer of the pollutant from bulk solution to the outer surface of the adsorbent, internal mass transfer from the outer surface to the inner pores of the adsorbent and the adsorption of adsorbate particles onto the active pores of the adsorbent. The overall rate of the reaction is determined by either film formation or intraparticle diffusion or both (Abdel-Raouf and Abdul-Raheim 2017). An ideal adsorbent for the adsorption of pollutants should include inexpensiveness, good mechanical and structural integrity to overcome water flow for a long time, high adsorption capacities with high rates, have a large surface area and possess a regeneration aptitude using cost-effective approaches (Mahfoudhi and Boufi 2017). Different materials have been tested as possible wastewater adsorbents such as polymeric, carbon-based, bio-based and inorganic materials. As per the above requirements, adsorption using low-cost materials with satisfactory adsorption properties and environmentally friendly nature has gained much attention and currently researchers have switched onto green adsorbents due to their abundance, biodegradability and non-toxic nature. Green adsorbents include low-cost materials originated from (1) natural sources (Sharma et al. 2011) (2) agricultural residues and wastes in particularly lignocellulosic biomass (Sud et al. 2008; Abdolali et al. 2014) and (3) low-cost sources (Bhatnagar and Sillanpää 2010) from which activated carbon adsorbents can be produced. Indeed green adsorbents were found to be inferior in terms of their adsorption capacity compared to commercial adsorbents such as activated carbons and structurally complex inorganic composite materials. But their cost-potential makes them competitive (Kyzas and Kostoglou 2014). Cellulosic adsorbents have the proficiency to meet almost all the requirement for being green and are thus potential materials for high end applications such as water purification.

Adsorption by green adsorbents

Interesting works have been reported regarding the adsorption of various heavy metals and dyes onto green adsorbents. Dried prickly pear cactus cladodes were explored for the biosorption of dyes from aqueous solutions (Barka et al. 2013). In another study, Ferrero explored the adsorption of methylene blue onto ground hazelnut shells and observed that adsorption capacities of methylene blue for hazelnut shells were five times higher than the respective amount reported for activated carbon obtained from the same material (Ferrero 2007). Animal bone meal was explored as a novel adsorbent for the removal of rhodamine B from wastewaters and the adsorption capacities obtained at different temperatures were close to 65 mg/g (El Haddad et al. 2016). El-Mekkawi and Galal investigated that the adsorption capacity of rutile TiO_2 and Degussa P25 TiO_2 for the removal of Direct Fast Blue B2RL and an adsorption

Table 2 Treatment technologies for the removal of pollutants from wastewater and associated advantages and disadvantages

Technology	Advantages	Disadvantages	References
Chemical precipitation	Simple operation, not pollutant selective, low capital cost	Sludge generation, sludge disposal cost, high maintenance costs	Aderhold et al. (1996)
Coagulation–flocculation	Bacterial inactivation capability, good sludge settling and dewatering characteristics, simple, economically feasible	Chemical consumption, increased sludge, high-cost sludge disposal problem	Aderhold et al. (1996)
Adsorption	Broad spectrum of pollutants, high capacity, fast kinetics, low cost, easy operating conditions	Performance depends on type of adsorbent, chemical derivatization to improve its sorption capacity	Ali and Gupta (2006)
Membrane filtration	Small space requirement, low waste, low chemical consumption	High initial cost, high maintenance and operation cost, membrane fouling, limited membrane lifetime	Qin et al. (2002)

capacity of 56 and 144 mg/g respectively was observed (El-Mekkawi and Galal 2013). In the light of the literature reviewed, the green adsorbents show higher adsorption ability for dyes than the metal ions. Table 3 depicts the adsorption capacities of various green adsorbents for the removal of dyes and heavy metals.

Adsorption on cellulose

Cellulose-based materials are abundant, cheap and have low or little economic value. Different forms of cellulosic materials are used as adsorbents such as fibers, leaves, roots, shells, barks, husks, stems and seed as well as other parts also. Jalali and Aboulghazi investigated the feasibility of sunflower stalks for lead (Pb) and cadmium (Cd) metal ion adsorption (Jalali and Aboulghazi 2013). Batch adsorption studies were conducted to study the effect of contact time, initial concentration, pH and adsorbent doses on the removal of Cd(II) and Pb(II) metal ions at room temperature. Elemental mercury adsorbents were successfully synthesized from the coconut husk wastes using different surface treatment methods (Johari et al. 2016). The surface morphology and surface functional groups of these adsorbents significantly changed after mercerization and bleaching treatments and resulted in different adsorption performances. The elemental mercury adsorption capacity for coconut pith and coconut fiber adsorbents observed the following trend: coconut pith-NaOH (956.282 mg/g) > pristine coconut pith (730.250 mg/g) > coconut fiber-NaOCl (639.948 mg/g) > coconut fiber-H_2O_2 (634.347 mg/g) > coconut fiber-NaOH (611.678 mg/g) > coconut fiber-H_2O_2 (531.277 mg/g) > coconut pith-NaOCl (501.126 mg/g > coconut fiber (431.773 mg/g). Table 4 shows the adsorption capacity of various lignocellulosic adsorbents, the major source of cellulose, for the removal of dyes and heavy metals. Most of the adsorption studies were conducted using untreated cellulosic materials and only a few of them show good adsorption potential. However, the performance of these adsorbents has been remarkably affected upon physical and chemical treatment. Table 4 shows the adsorption capacity of various

Table 3 Adsorption capacities of various green adsorbents for the removal of dyes and heavy metals

Green adsorbent	Dye/heavy metal	Adsorption capacity (mg/g)	References
Peat	Basic blue 3	555.61	Allen et al. (2004)
	Basic yellow 21	666.56	
	Basic red 22	312.50	
Bentonite	Malachite green	7.72	Tahir and Rauf (2006)
Chitin	Reactive yellow 2	2.83	Akkaya et al. (2007)
	Reactive black 5	1.88	
Hazelnut shells	Methylene blue	41.3	Ferrero (2007)
Palm ash	Begacron blue BBLS 200%	49.5	Isa et al. (2007)
	Miketon polyester scarlet RCS	61	
Treated olive stones	Cd(II)	49.3	Aziz et al. (2009)
Papaya seeds	Methylene blue	556	Hameed (2009a)
Musa spp	Violet 54	36.49	Kumar et al. (2010)
Cactus cladodes	Methylene blue, Eriochrome black T Alizarin S	189.83 200.22 118.35	Barka et al. (2013)
Rutile TiO_2	Direct fast blue B2RL	56	El-Mekkawi and Galal (2013)
Degussa P25 TiO_2	Direct fast blue B2RL	144	
Corn fibers	Alcian blue	159	Mallampati et al. (2015)
Luffa peals	Methylene blue	70	
	Neutral red	50	
	Coomassie brilliant blue	35	
	Alcian blue	90	
	Methylene blue	124	
	Neutral red	108	
	Coomassie brilliant blue	40	
Magnetic litchi peel	Pb(II)	78.74	Jiang et al. (2015)
Animal bone meal	Rhodamine B	65	El Haddad et al. (2016)
Citrus limetta peel	Methylene blue	227.3	Shakoor and Nasar (2016)
Grapefruit peelings	Mixture of dyes	37.427	Rosales et al. (2016)
	Cr(VI)	39.0628	

Table 4 Results on the removal of dyes and heavy metals by different cellulose-based adsorbent

Cellulose-based adsorbent	Dyes/heavy metals	Adsorption capacity (mg/g)	References
Garlic peel	Methylene blue	142.86	Hammered and Ahmad (2009)
Grass waste	Methylene blue	457.64	Hameed (2009b)
Jack fruit peel	Basic blue 9	285.71	Hameed (2009c)
Spent tea leaves	Methylene blue	300.05	Hameed (2009d)
Wheat straw	Cr(VI)	47.16	Dhir and Kumar (2010)
	Ni(II)	41.84	Dhir and Kumar (2010)
Rice husk	Direct red 31	74.07	Safa et al. (2011)
Sugarcane bagasse	Ni(II)	2.0	Alomá et al. (2012)
Mango seed	Victazol orange	44.8	Alencar et al. (2012)
Citrus waste	Reactive blue19	37.45	Asgher and Bhatti (2012)
Oil palm shell	Cu(II)	1.75	Chong et al. (2013)
	Pb(II)	3.39	
Sunflower stalk	Cd(II)	69.80	Jalali and Aboulghazi (2013)
Bamboo leaf powder	Hg(II)	27.11	Mondal et al. (2013)
Mosambi (sweet lime) peel	Cr(VI)	250	Saha et al. (2013)
Rice husk	Cr(III)	22.5	Sobhanardakani et al. (2013)
	Cu(II)	30.0	
Jute fiber	Congo red	8.116	Roy et al. (2013)
Cauliflower waste	Pb(II)	47.63	Hossain et al. (2014)
Pomegranate peel	Cu(II)	30.12	Ben-Ali et al. (2017)
Cucumber peel	Pb(II)	133.60	Basu et al. (2017)
Orange peel	Cu(II)	63	Guiza (2017)

lignocellulosic adsorbents, the major source of cellulose, for the removal of dyes and heavy metals.

Adsorption by modified cellulose

Cellulose is abundant in hydroxyl groups which can anchor other functionalities through a variety of chemical modifications. Modification of cellulose involves the direct modification and monomer grafting. Direct cellulose modification in the preparation of adsorbent materials is esterification, etherification, halogenation, oxidation, alkaline treatment and silynation (O'Connell et al. 2008; Hokkanen et al. 2016). Chemically modified cellulose bearing Schiff's base and carboxylic acid groups was synthesized for the removal of Cu(II) and Pb(II) from aqueous solutions (Saravanan and Ravikumar 2016). This novel green adsorbent was synthesized by periodate oxidation of cellulose followed by condensation reaction with p-aminobenzoic acid for the Schiff's base forming reaction. In another study, chemically modified cellulose bearing Schiff base extracted from Sesbania sesban plant was synthesized via a novel method using 2-hydroxy-5-methyl benzaldehyde for the removal of Cd(II) (Naeimi and Amini 2018). The cellulose biomass exhibited the highest metal ions uptake capacity of 9.39 mg/g at pH value of 4.0, biomass dosage of 0.01 g/L and cadmium concentration of 150 mg/L. Bediako et al. (2016) developed an adsorbent via carbomethylation and cross-linking reactions from waste lyocell fabric to produce carbomethyl cellulose adsorbent for the removal of Cd(II) (Bediako et al. 2016). This adsorbent displayed approximately 17 times greater metal uptake than the original material and at neutral and alkaline pH, maximum Cd(II) uptake was displayed (Table 5).

Adsorption by modified nano/microcellulose

Carboxycellulose nanofibers prepared from untreated Australian spinifex grass using a nitro-oxidation method were found to be an effective medium to remove Cd^{2+} ions from water. A low concentration of nanofiber suspension could remove large concentrations of Cd^{2+} ions in less than 5 min. The maximum Cd^{2+} removal capacity of the cellulose nanofibers was around 2550 mg/g and the highest removal efficiency of 84% was exhibited when the Cd^{2+} concentration was 250 ppm (Sharma et al. 2018a). Sun et al. (2017) prepared a cellulosic adsorbent by halogenation of microcrystalline cellulose followed by the functionalization with pyridone diacid for the removal of Pb(II) and Co(II) from aqueous solutions. The content of carboxyl groups in this cellulosic adsorbent was determined to be 1.32 mmol/g, which was responsible for the high adsorption toward metal ions. In another study, TEMPO-oxidized fibrous cellulose modified with polyethyleneimine via cross-linking with glutaraldehyde also exhibited a higher adsorption of Cu(II) at pH 5 than the polyethyleneimine grafted cellulose (Zhang

Table 5 Adsorption capacities of various modified cellulose adsorbents for the removal of heavy metals and dyes

Cellulose adsorbent	Modifying agents	Heavy metal/dye	Maximum adsorption capacity	References
Wood pulp	Succinic anhydride (carboxyl)	Cd(II)	169 mg/g	Geay et al. (2000)
Cellulose bead	1. Acrylonitrile 2. Sodium hydroxide (carboxyl)	Cr(III) Cu(II)	73.5 mg/g 70.5 mg/g	Liu et al. (2001)
Cellulose powder	Acrylic acid (carboxyl)	Pb(II) Cu(II) Cd(II)	55.9 mg/g 17.2 mg/g 30.3 mg/g	Güçlü et al. (2003)
Wood pulp	Citric acid (carboxyl)	Cu(II) Pb(II)	24 mg/g 83 mg/g	Low et al. (2004)
Cellulose (juniper fiber)	Sodium hydroxide (hydroxyl)	Cd(II)	0.26 mg/g	Min et al. (2004)
Cellulose powder	Acrylonitrile hydroxylamine (amidoxime)	Cu(II) Cr(III)	3.76 mmol/g 3.90 mmol/g	Saliba et al. (2005)
Cellulose pulp	1. Acrylic acid 2. Acrylamide carboxyl (amino)	Cu(II)	0.74 mmol/g	Bao-Xiu et al. (2006)
Wood sawdust cellulose	Sodium hydroxide (hydroxyl)	Cd(II)	0.65 mmol/g	Šćiban et al. (2006)
Sawdust cellulose	Sodium hydroxide (hydroxyl) untreated	Cd(II)	0.65 mmol/g	Memon et al. (2007)
Cellulose	Succinic anhydride (carboxyl)	Cu(II) Cd(II) Pb(II)	0.47 mmol/g 0.76 mmol/g 0.99 mmol/g	Gurgel et al. (2009)
Cellulose	Succinic anhydride + triethylenetetramine (carboxyl, amine)	Cr(VI)	0.82 mmol/g	Gurgel et al. (2009)
Cellulose	Acrylonitrile *N*,*N*-methylenebisacrylamide (amino)	Cd(II)	0.19 mmol/g	Zheng et al. (2010)
Cellulose	Succinic anhydride + sodium bicarbonate (carboxylate)	Co(II) Ni(II)	2.46 mmol/g 2.46 mmol/g	Melo et al. (2011)
Cellulose	Glycidylmethacrylate	Cr(VI)	2.38 mmol/g	Anirudhan et al. (2013)
Cellulose	Acrylic acid (carboxyl)	Cu(II) Ni(II)	5.17 mmol/g 4.71 mmol/g	Hajeeth et al. (2013)
Cellulose bagasse	HCl, HNO_3, NaOH tartaric, citric and oxalic acids (carboxyl)	Zn(II) Cd(II) Pb(II)	0.12 mmol/g 0.13 mmol/g 0.17 mmol/g	Velazquez-Jimenez et al. (2013)
Cellulose	Glycidyl methacrylate Diethylenetriamine acetic acid (carboxyl)	Malachite green Basic fuchsine	3.16 mmol/g 1.36 mmol/g	Zhou et al. (2013)
Cellulose	Methyl benzalaniline	Cu(II) Pb(II)	157.3 mg/g 153.5 mg/g	Saravanan and Ravikumar (2015)

et al. 2016). The functionalized nanocellulose obtained by selectively oxidizing the C_2 and C_3 hydroxyl groups followed by oxidizing the aldehyde groups to form 2, 3-dicarboxyl groups demonstrated a maximum adsorption capacity for Cu^{2+} at pH 4 (Sheikhi et al. 2015). Mautner et al. (2016) synthesized phosphorylated nanocellulose papers for copper adsorption from aqueous solutions. The nanopaper ion-exchangers were able to adsorb copper ions in dynamic filtration experiments on passing water containing copper ions through the nanopapers.

Organic dye pollutants display cationic, anionic or non-ionic properties and pose a significant environmental problem in many parts of the world. Cationic dyes are removed using nanocellulose functionalized with anionic moieties. Carboxylated nanocellulose synthesized via TEMPO-mediated oxidation resulted in a significantly higher uptake of 769 mg/g at pH 9 of the cationic dye methylene blue, compared to nanocellulose with sulfate groups on their surfaces with an adsorption capacity of 118 mg/g at pH = 9 (Batmaz et al. 2014). Carboxylated nanocellulose produced by citric acid/hydrochloric hydrolysis of microcrystalline cellulose was used for the adsorption of methylene blue (Yu et al. 2016). Anionic dyes are usually removed using nanocellulose functionalized with cationic moieties. Cationic nanocellulose prepared via successive sodium periodate oxidation followed by reaction with ethylenediamine displayed a maximum uptake of 556 mg/g of acid red GR (Jin et al. 2015a). The same functionalization method was explored by Zhu et al. on dialdehyde functionalized cellulose powder, but using hyper-branched polyethyleneimine. The adsorbent displayed a high Congo red adsorption of 2100 mg/g and a high cationic basic yellow adsorption of 1860 mg/g (Zhu et al. 2016).

Nanocellulose, in the form of carboxycellulose nanofibers prepared using nitro-oxidation method exhibited the highest adsorption capacity of 2550 mg/g for removal of Cd^{2+} ions from water. Among the dyes, carboxylated nanocellulose synthesized via TEMPO-mediated oxidation resulted in the maximum adsorption capacity of 769 mg/g for the cationic dye methylene blue. Table 6 shows the adsorption capacities of nano/microcellulose and their modified counterparts.

Table 6 Modified and unmodified nanocellulose adsorbents for the removal of heavy metals and dyes

Nano/microcellulose adsorbent	Modification	Heavy metal/dye	Maximum adsorption (mg/g)	References
Bacterial cellulose	Epichlorohydrin + diethylenetramine	Cu(II) Pb(II)	63 87	Shen et al. (2009)
Nanocellulose	TEMPO-oxidation	UO_2^{2+}	167	Ma et al. (2011)
Nanocellulose	Ammonium persulphate oxidation	Methylene blue	101	He et al. (2013)
Crystalline nanofiber	Glycidyl trimethyl ammonium chloride	Congo red Acid green25	664 683	Pei et al. (2013)
Nanocellulose	Xanthation	Cd(II)	154.26	Pillai et al. (2013)
Nanocellulose	Succinic anhydride	Pb(II) Cd(II)	367.6 259.7	Yu et al. (2013)
	Succinic anhydride + sodium bicarbonate	Pb(II) Cd(II	465.1 344.8	
Nanocellulose Nanocellulose	TEMPO oxidation –	Methylene blue Methylene blue	769 118	Batmaz et al. (2014)
Pristine crystalline nanocellulose	Sulfuric acid hydrolysis	Methylene blue	118	Batmaz et al. (2014)
Microfibrillated cellulose	Aminopropyltriethoxysilane	Ni(II) Cu(II) Cd(II)	2.734 3.150 4.195	Hokkanen et al. (2014)
Microcrystalline cellulose	Quaternary amine groups + ultrasonication	Congo red	304	Hu et al. (2014)
Nanocellulose	–	Ag(I)	34.4	Liu et al. (2014)
Crystalline nanocellulose	Sodium periodate oxidation + ethylene diammine	Acid red GR	556	Jin et al. (2015a)
Crystalline nanocellulose microgels	Polyvinyl amine	Acid red GR Congo red 4BS	896 1469	Jin et al. (2015b)
Nanocellulose	–	Ag(I) Cu(II) Fe(III)	120 114 73	Liu et al. (2015)
Nanofiber	Enzymatic phosphorylation	Ag(I) Cu(II) Fe(III)	136 117 115	
Nanocellulose	Phosphorylation	Ag(I) Cu(II) Fe(III)	136 117 115	Liu et al. (2015)
Pristine nanocellulose	–	Ag(I) Cu(II) Fe(III)	56 20 6.3	
Nanocellulose	Maleic anhydride	Crystal violet	244	Qiao et al. (2015)
Aminated nanocellulose	Sodium periodate + sodium alendronate	Vanadium	194	Sirviö et al. (2016)
Microcrystalline cellulose	–	Methylene blue	4.95	Tan et al. (2016)
Dialdehyde functionalized cellulose powder	Hyper-branched polyethyleneimine	Congo red Basic yellow	2100 1860	Zhu et al. (2016)
Microcrystalline cellulose	Halogenation + pyridine diacid	Pb(II) Co (II)	177.75 122.70	Sun et al. (2017)
Nanofiber	Nitro-oxidation	Cd(II)	2550	Sharma et al. (2018a)
Nanofiber	Nitro-oxidation	Pb(II)	2270	Sharma et al. (2018b)

Conclusion

In this review, applications of cellulose-based adsorbents for the removal of heavy metals and dyes from wastewater have been reviewed based on a substantial number of relevant research articles published up till now. Cellulose adsorbents are of specific significance owing to their abundant availability, ease of modification and application potential. As evident in the literature reviewed, most of the adsorption studies are limited to batch-scale only and are not fully developed at pilot and industrial scales for the treatment of real industrial effluents. Moreover, the actual industrial effluents were laden with several pollutants which require much work to investigate the selectivity of adsorbents in real effluents. Cellulose-based adsorbents contain many hydroxyl groups that can activate various reactions on chemical modification. Upon chemical modification, the adsorption capacity of these adsorbents has enhanced as a result of the increase in active binding sites on modification and addition of new functional groups that favor the higher uptake of pollutants. However, the pollution caused by the various modification methods was seldom reported. Currently, research is focused to synthesize modified cellulose and nanocellulose-based adsorbents using eco-friendly chemicals for wastewater treatment.

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