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Review Article

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Biomass Driven Nanocellulose: Latest Breakthrough in the Sustainable Adsorbent Materials Family for Confiscation of Water Toxicants

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Abstract

Owing to the copious existence of biomass in the environment, the present review is aimed at compiling some of the latest scientific reports focussed on the utilization of lignocellulosic biomass to fabricate environment friendly materials for wastewater remediation. Overall, this work is an attempt to probe proficient, inexpensive, eco-friendly nano dimensional materials based on biomass driven cellulose/nanocellulose or its modified forms that could be competently used as adsorbents for real wastewater treatment. The astonishing properties of cellulose have gained substantial attention from research consortium for its utility in wastewater decontamination as a bio-sorbent. The review encompasses information on distinct water toxicants, adsorptive removal of pollutants and research work going on in the field of wastewater remediation using bio-derived cellulosic materials as well as the hurdles faced by the scientific community in this field. Overall, it is perceptible that cellulose, being a biomass originated material, reduces the utilization of toxic chemicals, and provides an environment affable solution to the global trouble of wastewater disposal. Fabrication of such biomass based adsorbent materials can open new pavements for effective wastewater remediation.

Keywords

Biomass, Cellulose, Wastewater remediation, Nanocellulose, Adsorption

Introduction

Water is one of the most treasured gifts of nature to the humankind. But an unexpected increase in industrial discharges, agricultural practices and numerous urban activities have left this valuable resource polluted to a very brutal extent [1-3]. The ensuing impact of water contamination on human as well as aquatic life is afar an explanation. As a result of different agricultural, industrial, and domestic activities, various toxic contaminants (Figure 1) such as organic pollutants like coloured contaminants, pesticides, pharmaceutical waste i.e., drugs, heavy metal ions, radioactive pollutants and substituted aromatics are being relentlessly discharged into the water ecosystem which is a matter of great apprehension for the society [4, 5].

The aforementioned toxicants are known to cause health issues in human beings including cancer, mental abnormalities and other metabolic disorders [6-8]. They are also known to disturb the endocrine system of aquatic life. Accordingly, their abolition from wastewater prior to the water discharge into the water bodies is an obligatory task. The worldwide concerns being raised due to water pollution have prompted the scientific research community to find out sustainable and environment affable pathways for the eradication of such lethal toxicants from water resources [9].

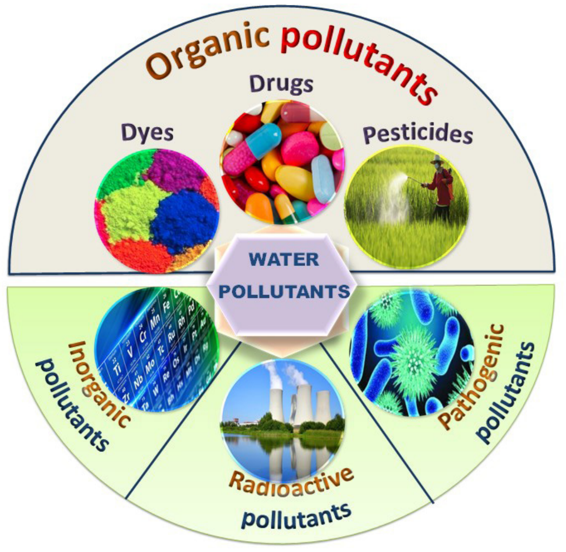

Figure 1: Different categories of water contaminants.

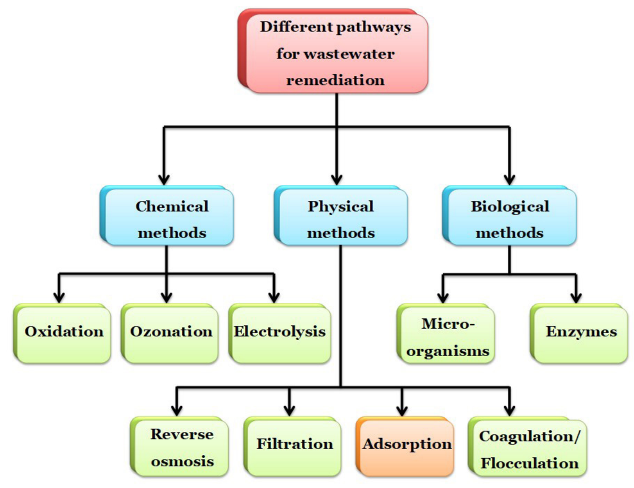

Figure 2: Different techniques for wastewater remediation.

Different Techniques for Wastewater Remediation

Till date, various methodologies have been exploited for the removal of wastewater contaminants and these can be largely categorized into chemical, physical and biological techniques as depicted in figure 2.

Although being broadly applicable, most of the physical treatment techniques exhibit some drawbacks [10-12] which includes high maintenance and operational cost, requirement of non-reusable coagulants and flocculants, high volume of sludge generation, high energy demand, complex instrumentation, low throughput, etc.

Therefore, a proficient and advanced technique which possess minimum disadvantages and can efficiently eliminate a broad range of recalcitrant toxicants from wastewater is required. To this problem, adsorption turns out to be the most apt solution owing to its numerous advantages as discussed ahead.

Adsorption: a simple yet efficient solution for wastewater remediation

Adsorption has outshined as an effective technique to salvage the deteriorating water resources due to hazardous pollutants. The efficacy of this technique lies in its versatility and operational simplicity for easy removal of distinct toxicants including inorganic pollutants, organic pollutants, and biological pollutants [13-15]. Besides being capable of removing different contaminants, adsorption method extends various other advantages as summarized in figure 3.

Figure 3: Advantages offered by adsorption technique.

Adsorption method of wastewater treatment has come out to be a better technique in comparison to other existing techniques like reverse osmosis, ozonation, coagulation, ion-exchange, ozonation, etc., because these techniques often involve higher operational cost as well as demand high capital investment. For this reason, these techniques are less practicable for large scale wastewater purification [16]. The simplistic set-up and designing of the adsorption technique combined with lesser requirement of initial investment in terms of capital and land area has made it one of the most potent techniques for wastewater indemnification [17].

Immense research has been executed so far for the elimination of pollutants from wastewater using different adsorbents. Activated charcoal was amongst the first and mostly utilized adsorbent in the early time but recently, several cost-effective adsorbents have substituted charcoal due to their environment promising characteristics. Adsorption technique has been widely utilized for the elimination of total dissolved solids (TDS) from industrial wastewater. As per literature reports, limestone aggregates serve as the potential adsorbent and removed 76.7% TDS [18]. Other adsorbents like aminated and sulfonated forms of bio-nanocomposites developed using chitosan and gum Arabic showed the removal of TDS up to 99.8% and 87.9%, respectively [19].

Moreover, Adsorption can be implied as a useful method to eliminate the toxicants present in municipal water supply. For instance, to remove organic pollutants like pesticides or pharmaceuticals, up to 90% or higher removal efficiency can be achieved [20], 80% - 99% of heavy metals can be removed

[21], inorganic impurities like fluoride and chloride can be removed up to 90% [22]. Therefore, it can be established that adsorption technique can be utilized as the most effective and convenient method of wastewater treatment.

Presently, nanotechnology has revolutionized the scientific community owing to the outstanding physical and mechanical properties exhibited by the nano-dimensional materials. Taking cognizance of the emerging benefits of nanotechnology merged with environment favourable materials, biomass derived nanomaterials have been recognised as one of the most outperforming adsorbents as described ahead.

Biomass: a key to address the world water challenges

The requirement of cheap, renewable and environment friendly materials is amongst the most common threads all the way through the history for the treatment of wastewater streams.

Currently, biomass stands as the most predominant resource for developing renewable and environment friendly materials around the globe and is acquiring considerable scientific attention among the research consortium aiming at sustainable development [23, 24]. Biomass is defined as the non-fossil form of carbon which is rich in energy and encompasses entire water- and land-based vegetation and incorporates the waste generated from municipal bio-solids, agricultural activities, forests, and industrial effluents [25].

Renewability being the most distinct advantage of biomass has made it a win-win solution to each environment related crisis. Another pronounced feature of biomass is its huge abundance and facile accessibility throughout the world. Biomass is easily available in all corners of earth in one way or the other all through the year. The additional benefit is its cost-effectiveness. Raw biomass is freely available in nature which can be facilely tailored into high performing adsorbent materials with minimum investment [26, 27].

Particularly, lignocellulosic biomass serves as a competent and promising feedstock for development of distinct adsorbent materials. Lignocellulosic biomass is practically inexhaustible because unless the sunlight remains available; the photosynthesis process will never come to a standstill; hence the lignocellulosic resources will never vanish. The major sectors which contribute towards lignocellulosic biomass are agriculture, industries, crop residues, forest, animal, and municipal residues, etc. The worldwide production of plant-based biomass, which includes 90% of lignocellulosic materials equals to roughly 200 x 10^9 tons per annum [28]. Particularly in a country like India, the budding interest in the lignocellulose biomass may be advocated to the increasing agricultural production of different crops like rice, corn, wheat, cotton, sorghum, sugarcane, etc., which produce large amount of waste which is mostly burnt and cause environmental hazards.

A deep insight into the structure of lignocellulosic biomass

To comprehend the intrinsic structure and the components which form lignocellulosic biomass, the thorough structure of plant cell wall must be understood. Lignocellulosic biomass is typically a miscellaneous material comprising of three different types of oxygen containing organic polymers i.e., lignin, hemicellulose, and cellulose. The major macro-components of any plant cell wall i.e., primary and secondary cell wall, middle lamella as well as the plasma membrane comprises of these three principal components [29]. An exemplary representation of plant cell wall and the constituent materials is demonstrated as figure 4.

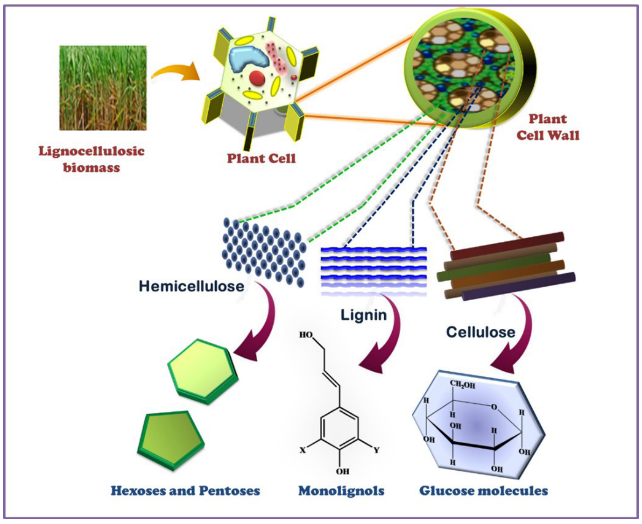

Figure 4: An insight of plant cell wall containing hemicellulose, lignin, and cellulose.

Cellulose: a versatile moiety with huge potential

Among all the components of lignocellulosic biomass, cellulose is a fascinating biopolymer and a focus of extensive research in the field of wastewater remediation. A French chemist, Payen [30] discovered cellulose in 1838 and since then, cellulose has been recognised to be a saviour to fulfil the dire need of renewable and biodegradable materials for the safety of environment. Many expectations have been set forth on cellulosic materials as compared to other lignocellulosic biomass materials on account of their exceptional physical and chemical properties for instance mechanical toughness, biodegradability, chirality, stereoregularity, biocompatibility, large and active surface area, hydrophilicity, lower cost, and higher capacity of undergoing chemical modifications [31-33].

In structural aspects, cellulose is a linear polysaccharide having high molecular weight and chemically formed from β-D-anhydro glucopyranose units. The molecular formulation of cellulose is expressed as $(C_6H_{12}O_6)_n$; wherein n denotes the degree of polymerization. The glucopyranose units bond together through a covalently bonded oxygen atom bonded to C-1 of one glucose unit and C-4 of the adjoining glucose unit. This linkage is called as β-(1-4) glycosidic linkage as indicated in figure 5. One end of the cellulose chain consists of one free anomeric carbon and is therefore called the reducing end whilst the other end has no free anomeric carbon atom and is termed as the non-reducing end.

The intrinsic cellulose structure varies strongly depending on the source from which it is extracted. Some of the sources from agro-industrial sector and forest residues have been outlined in table 1. It is progressively being more realized that

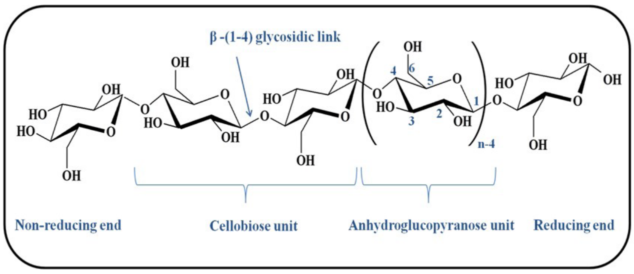

Figure 5: Cellulose chain demonstrating the chair conformation of anhydroglucose unit, glycosidic linkage, and the reducing and non-reducing ending points of cellulose polymer.

Table 1: Varying percentage of cellulose present in distinct agro-industrial and forest residues.

Biomass	Cellulose (%)
Rice straw	28 - 36 [34]
Wheat straw	33 - 38 [35]
Sugarcane bagasse	40 - 50 [36]
Sorghum bagasse	34 - 45 [28]
Barley straw	31 - 45 [28]
Oat straw	31 - 37 [37]
Rice husk	25 - 35 [35]
Softwood	42 - 50 [36]
Hardwood	39 - 48 [36]
Corn cob	33 - 42 [36]
Corn leaves	25 - 30 [35]
Bamboo	38 - 42 [35]
Switchgrass	5 - 20 [38]
Nutshells	25 - 30 [38]

these residues can act as key precursor materials for the fabrication of distinct cellulose-based products.

Nanocellulose: a journey from biomass waste to sustainable high-performance adsorbent

Dates back, the prominence of cellulose was extremely accepted but it was the arrival of nanotechnology which took this biomaterial to further heights. It has been unveiled in literature that by extracting cellulose at nanoscale, most of the defects and amorphous regions present in the hierarchical structure of cellulose can be removed, leading to a new generation of cellulose based materials [40-42]. Based on the dimensions, morphology and the method of processing, there are four forms of nanocellulose viz cellulose nanocrystals (CNC) [43], cellulose nanofibers (CNF) [44], bacterial nanocellulose [45] and microfibrillar cellulose [46]. The surface of nanocellulose can be readily tuned by either grafting some organic or inorganic moieties on the surface or by composting it with different moieties to enhance its native properties. Various research cohorts around the globe have put their best efforts to utilize these modified forms of cellulose for indemnification of different water toxicants as discussed ahead.

Liu et al. [47] developed a multifunctional aerogel based on biomass using chitosan and cellulose extracted from waste reed. The aerogel possessed abundantly porous three-dimensional structure with a very low density. The prepared aerogel was employed for the simultaneous adsorptive removal of

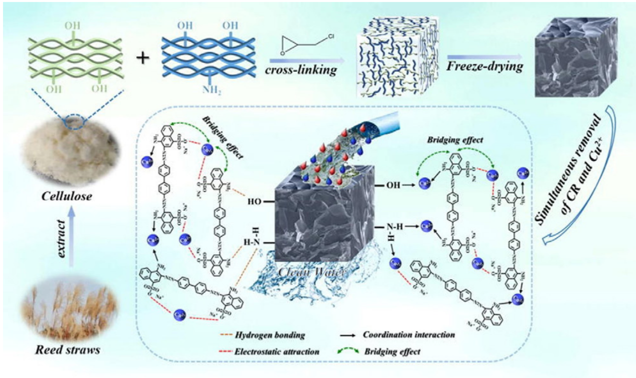

Figure 6: Removal mechanism of Congo red and Cu^{2+} using cellulose-chitosan aerogel [41].

Congo red and cupric (Cu^{2+}) ions from wastewater having maximum adsorption capacity of 380.23 mg/g and 260.41 mg/g, respectively. The authors suggested that the preloaded Congo red molecules offered new adsorption sites for the facile bridging of free Cu^{2+} present in the solution. In a similar manner, preabsorbed Cu^{2+} ions provided the bridging effect for Congo red dye molecules as depicted in figure 6.

Vijayan and Prabhu [48] prepared nanocellulose and carboxylated nanocellulose from two different biomass sources i.e., *Desmostachya bipinnata* and Hybrid Napier CO5 grass. The authors used peroxide bleaching method for the extraction of nanocellulose. Maghemite was used as a catalyst for the depolymerization of cellulose and conversion into nanocellulose. The prepared nanocellulose and its carboxylated form were utilized as adsorbents for the removal of Evans blue dye. Both the adsorbents were found to be reusable with excellent adsorption capacities.

Khadir et al. [49] used nanoparticles of conducting polymeric polypyrrole-polyaniline system to modify sisal cellulosic fiber. The authors utilized it as a cost effective and non-toxic bio-nanocomposite for the adsorptive elimination of ibuprofen. A very high removal efficacy of 88% was attained at pH 5 in 60 min at an adsorbent dosage of 150 mg. It was revealed from the energy site distribution analysis that more adsorbent sites were active at high temperature leading to higher adsorption capability with the maximum removal capacity of 19.45 mg/g, respectively.

Bhatti et al. [50] meticulously studied the adsorption of different dyes i.e., Direct Red-31, Ever Direct Orange-3GL, Direct Orange 26, Direct Blue-67 onto the surface of modified rice husk, carboxymethyl cellulose, alginate and palginate, alcohol immobilized biomasses. Fourier transform infrared (FTIR) spectroscopy analysis was performed to study the role of hydroxyl, carboxylic acid, and amino groups in the adhesion of dye molecules onto the biomass surface. The authors concluded that modified rice husk exhibited excellent potential for the degradation of textile effluents.

Bisla et al. [51] performed the extraction of CNF from rice straw through soda cooking methodology leading to the hydrolysis of hemicellulose components and yielding pure nanofibers of cellulose. The resulting nanofibers were functionalized with L-methionine to graft sulfide and amino groups on the fiber surface. The functionalized fibers exhibited an adsorption efficiency of 131.86 mg/g for the adsorptive uptake of Hg(II) ions at a pH = 7.8 and room temperature. The mechanism of adsorption be fitted well with pseudo-second-order kinetics and Langmuir isotherm model of adsorption.

α-cellulose fibers were sourced from wastepaper biomass (WP-αCFs) by Kadam et al. [52]. The fibers were magnetized with Fe_3O_4 and the prepared biosorbent was utilized for the remediation of cobalt oxide nanoparticles. The adsorption mechanism was found to follow pseudo-second-order kinetics. Langmuir isotherm model was found to sit well with the obtained adsorption data demonstrating an extremely high adsorption capacity of 1567 mg/g. Thus, wastepaper derived bio-nanocomposite having an extraordinary adsorption potential was established as an efficacious tool for wastewater remediation.

Pourfadakari et al. [53] scrutinized the efficiency of nanocellulose extracted from rice husk for the adsorptive elimination of Cr(VI) ions from wastewater. All the adsorption related parameters were optimized, and the best adsorption results were achieved at an adsorbent dosage of 1.5 g/L at pH = 6 and an adsorbent-adsorbate contact time of 100 min at pollutant concentration of 30 mg/L. Additionally, the experimental data was found to corelate well with Langmuir isotherm model and the adsorption reaction followed pseudo-first-order kinetics. From thermodynamic studies, it was revealed that Cr(VI) adsorption onto nanocellulose surface was a spontaneous and endothermic process.

Rani et al. [43] utilized steam explosion methodology for the extraction of CNCs from banana fibres. After the extraction procedure, ceric ammonium nitrate was used as an initiator to carry out the polymerization reaction to graft butyl acrylate monomer on the surface of CNCs as depicted in figure 7. Efficient synthesis of grafted copolymer was proven from FTIR analysis. Heterogeneity of the surface of grafted copolymer was confirmed from scanning electron microscopy images which aided in facile adsorption of Pb(II) ions. Maximum Pb(II) elimination was observed at pH 5 with a polymer dosage of 4 g per 100 ml, adsorption contact time of 360 min and initial concentration of 125 mg/L.

Sehaqui et al. [54] worked on the preparation of cationic CNF out of waste pulp residues by introducing positively charged quaternary ammonium groups in different amounts. Initially, waste pulp was exposed to etherification by treating with glycidyltrimethylammonium chloride followed by mechanical disintegration. The amount of positively charged ions was scrutinized using zeta-potential studies. Etherified

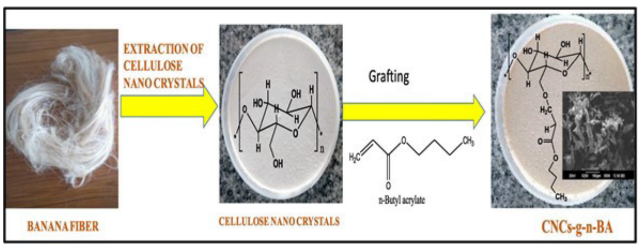

Figure 7: Grafting of butyl acrylate on CNCs [48].

CNF were used as an adsorbent for the removal of negatively charged toxicants i.e., nitrate, sulphate, fluoride, and phosphate ions. Authors observed that that the cationic CNF could successfully adsorb multivalent ions (sulphate and nitrate ions) with much more efficacy in comparison to monovalent anions (fluoride and nitrate ions).

Gupta et al. [55] combined differently modified forms of graphene oxide with esterified CNF sourced from sugarcane bagasse via steam explosion technique. The prepared adsorbents were tested for their adsorption efficacy towards pharmaceutical toxicants ciprofloxacin and ofloxacin. It was observed that the adsorption potential of the modified CNF was considerably ameliorated on blending with functionalized graphene oxide. Also, it was concluded by the authors that at 20% weight wise loading of carboxylated graphene oxide in esterified CNF, maximum removal of both the contaminants occurred.

Kankılıç and Metin [56] used chemical extraction process to produce cellulose microfibrils from *Phragmites australis*. The applicability of these microfibrils was investigated for the removal of methylene blue dye. A maximum adsorption capacity of 54.9 mg/g was recorded, and the adsorbent was found to be applicable over a broad range of pH. Experimental data satisfied both Freundlich and Langmuir models of adsorption thus indicating multilayer nature of adsorption.

Nguyen et al. [57] formulated a composite aerogel by a combination of environmentally friendly CNF extracted from nipa palm tree and graphene oxide via simple solvent evaporation method. The fabricated aerogel was found to exhibit porosity of more than 98.2%. The aerogel could rapidly and effectually update methylene blue dye from aqueous solution, with a 99% uptake in 20 min. The adsorption of methylene blue over the aerogel occurred through electrostatic interactions and the data agreed well with the pseudo-second order kinetic model.

Kara et al. [58] utilized *Eichhornia crassipes* biomass for the preparation of nanocellulose which was further modified using sodium periodate. The developed adsorbent was used to remove cationic methylene blue dye from its aqueous solution. A maximum removal capacity of 90.9 mg/g was achieved with removal percentage of 78% in 60 min at 30 mg/L concentration of the dye. Adsorption data was well fitted with pseudo-second order kinetic model and the adsorption process was observed to be feasible and spontaneous from thermodynamic studies.

Baruah et al. [59] synthesized a bio-nanocomposite of iron oxide and nanocellulose driven from sugarcane bagasse and rice husk. The nanocomposite was found to be superparamagnetic in nature and was therefore magnetically recoverable. It was proposed by the authors that the developed nanocomposite was able to remove almost 99% of arsenic from wastewater via chemisorption mechanism. Furthermore, the adsorbents were recyclable and more efficient than the commercially available adsorbents.

In another study, the synthesis of amide functionalized cellulose was proposed by the cross-linking interaction between biacrylamide and cellulose by Liu et al. [60]. The adsorption potential of the fabricated material was investigated using two i.e., dyes acid red 18 (AR18) and acid black 1 (AB1) and Cu^{2+} ions as model pollutants. The q_{max} for AR18, AB1 and Cu^{2+} ions were 751.80, 417.90 and 51.30 mg/g, respectively. The adsorptive removal of anionic dyes was increased due to the strong electrostatic forces of interaction between the amide groups on the adsorbent and SO_3^{2-} groups on the dyes (Figure 8).

Recently, Li et al. [61] worked in the fabrication of a biomass-based aerogel consisting of a combination of TEMPO-oxidized nanocellulose, starch, calcium carbonate nanoparticles and polyacrylamide. The aerogel exhibited a three-dimensional porous structure which was found to be useful for the effective adsorptive uptake of Congo red and methylene blue dyes with an adsorption capacity of 277.76 mg/g and 101.01 mg/g, respectively.

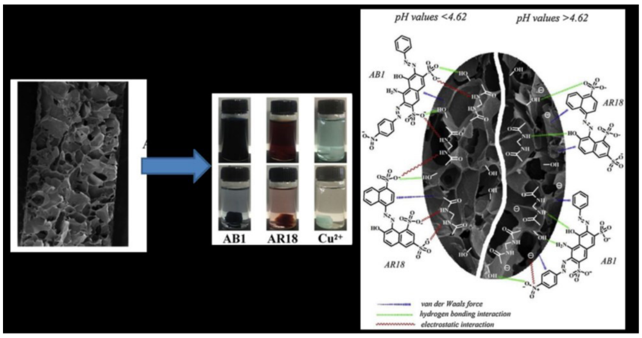

Figure 8: Adsorption mechanism between amine functionalized cellulose and dyes [55].

Conclusions and Future Perspective

The above-mentioned literature markedly depicts that biomass originated cellulose has come-forth as a sustainable and potential candidate to abolish noxious pollutants from wastewater. It can be employed as a bio-sorbent in versatile manners i.e., in its native form, in grafted/functionalized form or in combination with other reagent materials. Various cellulose-based platforms discussed in this review offer evidence that treatment of water at laboratory scale is feasible, but to check for the industrial scale application of the significant number of research reports is still a challenge. To accelerate the industrialization process of using biomass based cellulosic materials, stakeholders in the field of water management and membrane technology must undertake detailed guidelines on research and design (R & D), application and industrialization stage.

To fully unlock the potential of cellulose derived from biomass, it is requisite to engineer more economic bio-nano-systems which can promptly and efficiently contribute to eliminate the emerging wastewater toxicants and hence assist as environment saviours. Besides being inexpensive and environment affable, more notably, these platforms may also offer practicable solutions to handle different challenges of off-grid potable water which are faced by underdeveloped countries like India, as well as the treatment of wastewater from various industries.

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None.

Conflict of Interest

None.

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