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Review

Sustainable Nanotechnology Based Techniques for Mitigating the Pollutants from Pulp and Paper Industry

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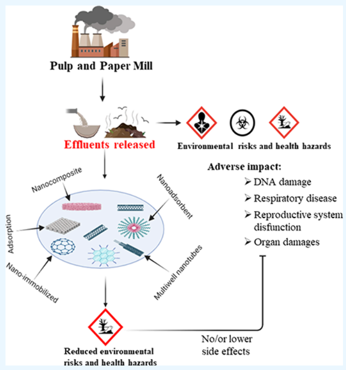
ABSTRACT: Paper mills inevitably produce various pollutants, including chlorolignin, chlorophenols, chloroguaiacol, furan, cyanide, and heavy metals. These pollutants cause significant threats to aquatic and terrestrial life. The pulp and paper industries are looking for eco-friendly solutions for the disposal of effluents during paper processing. Moreover, environmental management practices are a key concern that may be addressed by removing these effluents using suitable bioremediation techniques. Therefore, we have discussed several eco-friendly nanotechnology based sustainable bioremediation technologies like the use of nanoparticles, nanomaterials, nanocomposites, nanoadsorbents, and several advanced methods such as electrocoagulation and photocatalysis, which may be utilized for the elimination of hazardous pollutants from paper industry effluents. This review finally includes critical insight into the potential use of the above-mentioned nanotechnology based interventions for mitigation of contaminants from the paper industry. Nevertheless, there are a few limitations and challenges toward implementation of such technologies, which are also discussed in this review.



1. INTRODUCTION

The paper manufacturing industry is a rapidly growing sector worldwide. Even in this era of the digital world, paper has its significance. To meet the massive demand for paper, its production is done on a large scale in industries. Industrialization has pros and cons as it leads to many environmental issues, deterioration of human health, and marine diversity.[1] Effluents produced by industries consist of many toxic substances generated during industrial processes. The sludges and wastewater are the main form of pollutants which is generated from paper industries.[2] These are harmful to wildlife and marine life and have an adverse effect, causing respiratory issues and mutagenic effects.[3] A high amount of wastewater is released from paper industries during processes such as wood debarking, washing pulp, chemical bleaching, and paper-making.[4] Wastewater includes a variety of pollutants like dyes, chlorinated compounds, chlorinated hydrocarbons, chlorolignin, heavy metals, tannin acid, lignosulfonic acids along with various other organic and inorganic salts that have an adverse effect on the environment and public health.[5−7] Bleach effluents generated after the bleaching process contain chlorinated compounds that are recalcitrant and accumulated in the water ecosystem, inhibiting the food chain of aquatic life.[8] Thus, the industrial organic pollutants are straight contaminating source for the water reservoirs by enhancing the biochemical oxygen demand (BOD), chemical oxygen demand (COD) and total organic carbon (TOC) which has severe environmental and human health issues.[9,10] Therefore, it is essential to treat wastewater before releasing it into water bodies. Treatment of effluents generally means the removal of the TOC, dark color, and reduction in COD and BOD.[11,12] Various conventional methods, like sedimentation, flocculation, precipitation, filtration, and ion exchange, are used for the remediation process. Nonetheless, these procedures have many drawbacks including needs huge areas, being less efficient and expensive, producing additional harmful substances, and requiring a lot of energy.[13,14]

Consequently, there is a necessity of sustainable and cost-effective treatment technologies that have high capability, less cost, higher efficiency, and low energy consumption and can be easily operated.[15] Nanotechnology approaches have emerged as a practical discipline in eliminating effluents from the paper industry as a result of development and improvement in the nanobiotechnological sector.[16] Nanotechnology is advantageous as (i) it requires less area and less energy consumption and works rapidly, (ii) it has nascent features and processes,

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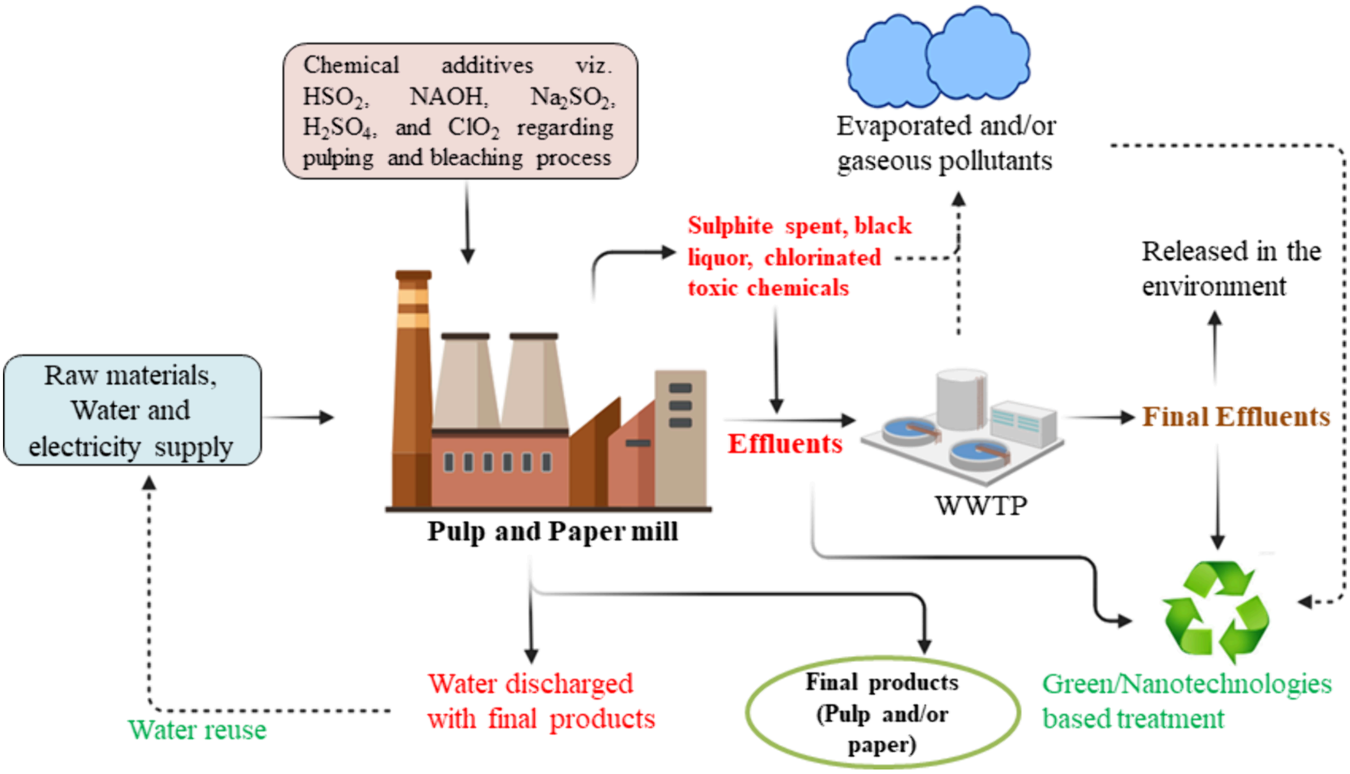

Figure 1. A schematic illustration of hazardous effluents discharged from a typical Pulp and Paper mill and their management through wastewater treatment plant and emerging green technologies.

and (iii) it has an effective length scale for production.[17] For instance, various nanocatalyst and nanoadsorbents like ZnO-NS, Co-NS, Au-NS, and Ag-NS have significant nanocatalyst properties for wastewater treatment. However, the capability of these nanostructures has been further enhanced with the support of cellulosic organic polymers.[18−20] Additionally, porous NS such as Cu_2SnS_3, CuO nanofibers, SiO_2 nanowires and nanorods also have efficient removal of industrial dyes.[21−23] According to Maqbool et al. titanium doped activated carbon−cellulose nanocomposite has significant and efficient rapid removal of organic pollutants other than conventional treatment processes.[23] The current review summarizes various nanotechnology based applications, such as using nanoparticles, nanocomposites, and nanoadsorbent technology for removing pollutants/hazardous elements from the effluent of pulp and paper industries. Moreover, some advanced methods, such as electrocoagulation, photocatalysis, and adsorption by carbon and polymer resins, are also discussed. This review also aims to provide an efficient understanding of nanotechnological applications in removing pollutants from pulp and paper mill effluents.

2. PULP AND PAPER MILL EFFLUENTS

In the paper manufacturing process, during pulping, bleaching, and washing steps, wastewater is released. In addition to this, other pollutants that are produced from the pulp industry include thermal loads, sizing agents, and coliform groups, such as turpentine and microbial biomass. During the process of pulping, a dark brown color liquid is generated, known as black liquor.[24] The presence of dissolved opaque compounds and their derived outputs, such as resins, and phenolic and acidic components, is responsible for the dark brown color.[25] Chlorides, chelating agents, transition metals, and dioxins are found in paper mill waste that has a significant impact on the ecosystem. The emission of sulfur-reducing compounds in a gaseous form, such as hydrogen sulfide, dimethyl sulfide, nitrogen oxide (NO_x), sulfur dioxide (SO_2) and particulate matter, causes air pollution.

Moreover, chlorinated compounds released into the water bodies during the bleaching stage react with organic compounds and form organochlorine compounds.[26] During paper production, wood extracts containing lipophilic components (sterol esters and triglycerides) and hydrophilic substances (lignin-like substances, low-molecular-mass lignin, and hemicellulose) are used. Waxes and lipids are removed from the wood with the help of organic solvents such as acetone and diethyl ether. The alkylphenol ethoxylate (APEOs) and linear alkylbenzenesulfonate (LASs) surfactants are found in the wastewater discharged from the pulp and paper mill. APEOs are nonionic surfactants, whereas LASs are anionic surfactants. APEOs are harmful to the water ecosystem and affect animals by stimulating an endocrine disturbance.[27] During the pulp bleaching process, chlorinated phenols are generated by the breakdown of chlorobenzenes. Chlorine compounds are crystal-like solids at ambient temperature. These monomeric chlorophenol compounds comprise mono-, di-, tri-, and tetrachloro isomers and pentachlorophenol, except liquid *o*-chlorophenol.[28]

Biocides are employed in the paper manufacturing industries to inhibit microbial, algal, and fungal growth. Their classification is based on mechanisms such as membrane-active biocides, cytotoxic agents, cell wall inhibitors, and genotoxic compounds. Excessive biocides in white water can reduce the effectiveness of secondary treatment. Two types of biocides are primarily used in various paper mills. One class includes oxidizing agents (hydrogen peroxides and chlorine

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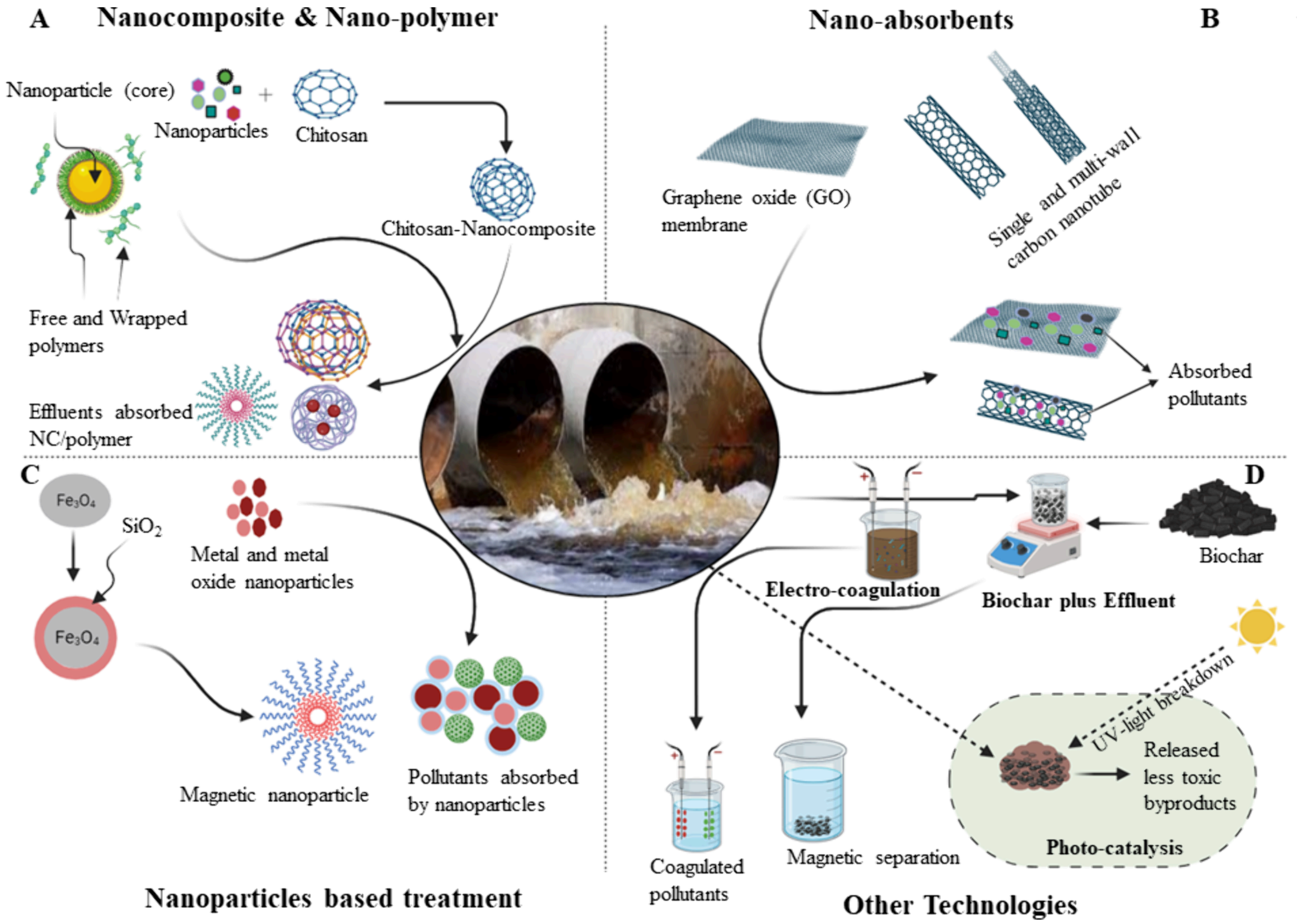

Figure 2. Effects and mechanisms of various ecofriendly and sustainable nanotechnological based techniques on remediation of hazardous effluent: (A) functionalized nanocomposite/polymers based treatment approach, (B) absorption of organic contaminants via nanoabsorbents, (C) nanoparticle based remediation of contaminants, and (D) simultaneous adsorption and degradation of contaminants through emerging technologies.

dioxides) used in pulp bleaching. These oxidizing agents either kill bacteria and fungus or weaken their cell walls, rendering them more sensitive to other biocides.[29] The other class of biocides contains extremely poisonous organic compounds (isothiazolinone, dithiocarbamate, thiocyanates, cyclobutane, and bromo compounds), which are used to preserve wood.[30] Another category can prevent biofilm development, e.g., surfactants such as alkyl sulfosuccinates. The effluents released from the paper mill at different stages are illustrated in Figure 1.

Paper manufacturing industries are considered substantial polluting industries due to the release of inorganic metallic and nonmetallic effluents in the composite nature of the aquatic system.[31] Ferrous, cadmium, copper, chromium, manganese, and nickel are the metallic compounds and sulfate, phosphate, potassium, chlorine, sodium, and magnesium are the non-metallic compounds. Arsenic generated in the industrial process of the papermaking industry is known to inhibit plant growth.[29] The inorganic structures of arsenic are arsenide and arsenate, which cause health hazards and are highly toxic to aquatic life. These are carcinogenic to human health, causing skin cancer and lung cancer. Metal-mediated free radicals cause different alterations in DNA bases, enhance lipid peroxidation, and disturb calcium homeostasis as well as sulfhydryl homeostasis.

3. NANOTECHNOLOGY BASED TECHNIQUES FOR EFFLUENT REMOVAL

3.1. Use of Nanoparticles. Nanoparticles are used in dechlorination, desalination, and removal of harmful substances like heavy metals and organic and inorganic substances from wastewater. Nanoparticles are metal-oxide and metal nanoparticles. The overall diagrammatic summary of various eco-friendly and sustainable nanotechnology based techniques are shown in Figure 2.

3.1.1. Metal-Oxide Nanoparticles. Various metal-oxide nanoparticles, i.e., magnesium oxide (MgO), zinc oxide (ZnO), titanium oxide (TiO_2), and ceric oxide (CeO_2), and other oxides used for adsorption of organic dyes have aroused researcher's curiosity because they are chemically inert, eco-friendly, and highly porous and can be easily synthesized.[32] Microwave-aided combustion synthesis and coprecipitation made ZnO, MgO, TiO_2 and CeO_2 nanoparticles. The characterization of these nanoparticles was done using the X-ray diffraction technique and field emission scanning electron microscopy.[33−35] Several researchers used these nanoparticles to remove different dyes, and their results depict favorable outcomes. Priyadarshini et al. studied the adsorption of Xylenol Orange dye using ZnO nanoparticles. Thermodynamic variables such as $\Delta H°$ disclose that the reaction is exothermic as it possesses a negative value (−0.115 kJ/mol), and $\Delta S°$

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Table 1. Various Nanotechnological Applications for Treatment of Pollutants with Removal Efficiency

Type of Material	Properties	Pollutant type	Operational parameter	Performance	Reference
Nanoparticles/Adsorbents					
TiO_2	10–50 nm of particle size	Zn(II), Cd(II)		Capacity: Zn (15.3 mg/g) and Cd (7.9 mg/g)	38
Hematite (a-Fe_2O_3)	Granular, with 75 nm crystal size	Cu(II)	25 °C and (pH 5.2 ± 0.1)	84.46 mg/g	39
CeO_2	260 nm size of hollow nanospheres	Cr(VI) Pb(II)	Ambient temperature	Absorbed 15.4 mg/g (Cr) and 9.2 mg/g (Pb)	40
	6.5–12 nm of particle size	Cr(VI)	Ambient temperature	Sorbed 121.95 mg/g of Cr (VI)	41
ZnO	<100 nm particle size	Zn Pb Cu	Mean temperature of 16.4 ± 1.8 °C and 41.4 ± 1.4% relative humidity	Increased retention capacity by Zn (73%), Cu (66%) and Pb (>99%) from stormwater	42
CMCD-MNPs coated with Fe_3O_4	12 nm	Cu ions	At 25 °C and 30 min of equilibrium	Highest adsorption of Cu ions: 47.2 mg/g	43
Ag@MSA (mercaptosuccinic Acid)	Silver nanoparticles with 9 ± 2 and 20 ± 5 nm	Hg(II)	At 1:6 ratio of Ag with MSA	Had a better removal capacity: 800 mg/g	44
Activated Carbons					
Activated carbon	With 486 m^2/g surface area	Cr(IV)	At pH 10.0, reaction time 180 min and temperature 298 K	Sorption capacity: 3.46 mmol/g	45
Granular activated carbon	Granular	As(V)	The optimal operational parameters were pH 2–11.5, 72 h of reaction time and temperature 283–328 K.	The highest sorption capacity: 2.5 mmol/g	46
Carbon Nanotubes					
MWCNTs-ox	Multiwalled with surface area of 197 m^2/g	Pb(II)	At 6 pH, reaction time of 1 h and 293 K temperature	The maximum adsorption capacity of 9.92 mmol/g of Lead.	47
CEMNPs	Surface area: 6–11 m^2/g	Cu(II)	pH 5–6 and 4 h equilibrium time	Adsorption capacity: 3.21 mmol/g	48
CNTs		Pb(II)	Optimum conditions are pH 3–7, reaction time of 6 h and temperature of 298 K	Adsorption capacity: 7.5 mmol/g	49
Graphene and Graphene Oxide					
Functionalized graphene oxide (FGO)	Surface area: 3.8 m^2/g	Co(II) Cd(II) Pb(II) U(IV)	At pH 6–9 and temperature of 293–333 K	Maximum adsorption capacity for Co(II) (106.3 mmol/g), Cd(II) (68.2 mmol/g), Pb(II) (842 mmol/g), and U(II) (97.5 mmol/g) reported	50–52
Poly(amidoamine) modified GO		Hg(II)	At room temperature and 24 h of contact time	Adsorption capacity: 0.5312 mmol/g	53
EDTA-GO	Surface area: 623 m^2/g	Pb(II)	At pH 6.8, reaction time 10–30 min and 323 K temperature	Adsorption capacity: 479 mmol/g	54

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Table 2. Application of Nanocomposite Materials in the Treatment of Hazardous Pollutants

Biochar composite type	Synthesis/feedstock	Characteristics	Technique used and conditions	Pollutant	Performance	Reference
Nanometal oxide/hydroxide	Tomato plant tissue	Presence of nanoscale $Mg(OH)_2$ and MgO particles	Pyrolysis at 600 °C up to 1 h	Phosphate	Mg enriched biochar removed 88.5% percent of phosphate from the solution.	80
	Cotton Wood	Biochar/AlOOH nanocomposite with carbon-nanoparticle structure	Pyrolysis $AlCl_3$-pretreated biomass at 600 °C up to 1 h	Arsenic, methylene blue and phosphate	Excellent adsorption capacity of biochar/AlOOH nanocomposite: Arsenic: 17410 mg/kg Methylene blue: 85000 mg/kg Phosphate: 135000 mg/kg	81
	Lotus stalks	Enhanced biochar yield and promoted biochar surface	Pyrolysis of zinc borate-pretreated biomass at 300 °C, 350 °C, and 400 °C up to 1 h	Ni(II)	Improved 3−10 times higher removal ability than pristine biochar	82
	Sewage sludge	Obtained more micropores and mesopores with higher surface area using fabricating agent	One step pyrolytic process in-combination with mixed fabricating pore agent (citric acid-$ZnCl_2$) at 500 °C up to 1 h	Benzene derivatives	Significantly sorb the all four benzene derivatives such as phenol (0.023 mmol/g), 4-chlorophenol (0.025 mmol/g), benzoic acid (0.052 mmol/g) and 4-hydroxybenzoic acid (0.058 mmol/g) from the aqueous solution	83
	Pecan nut shells	Improved the micropore and the total pore volume	Pyrolytic process of calcium salt-pretreated biomass at 600 °C up to 4 h	Dye (reactive blue 4 and acid blue 74)	It has higher removal efficiency (around 10 mg/g of sorbent) than pristine biochar.	84
Magnetic biochar	Empty fruit bunch	Magnetic biochar contains high 890 m^2/g surface area	Microwave assisted process using ferric chloride hexahydrate	Methylene blue	265 mg/g of sorption capacity with 99.9% efficiency	85
	Orange peel	Amorphous biochar and nanosize particles	Fe^{3+}/Fe^{2+} coprecipitation on orange peel powder by chemical process	HOCs and phosphate	Maximum sorption ability for phosphate (2.4 mg(P)/L) with 99.4% efficiency by magnetic biochar than the nonmagnetic biochar	86
	Rice husk and municipal solid waste organic fraction	Impregnated of biomass with iron and calcium agents before pyrolysis and pyrolytic process done at 300 °C for 1 h	Fe and Ca content increased	As(V)	Obtained more than 95% As(V) removal efficiency as compared to the nonimpregnated biochar	87
	Pyrolysis and then Precipitation	Fe_3O_4/Douglas fir biochar	Pyrolysis	Emerging contaminants such as caffeine, ibuprofen and acetylsalicylic acid	Enhanced adsorption capacity of adsorbent with 75.1 ± 1.8 mg/g, 39.9 ± 1.2 mg/g and 149.9 ± 4.5 mg/g for caffeine, ibuprofen and acetylsalicylic acid, respectively	88
Functional nanoparticles coated biochar	Wheat straw	More functional group and greater thermal stability	Slow pyrolysis of graphene pretreated biomass at 600 °C up to 1 h	Phenanthrene	Removal capacity increased nearly 30.4% at doses of 5−8.5 mg biochar. 100% removal efficiency was obtained at 30 mg of biochar.	89
	Bamboo, sugar cane bagasse, hickory wood and peanut hull	Biochar surface coated with chitosan with additional amine groups	Pyrolysis was done at 600 °C up to 2 h	Pb(II), Cu(II) and Cd(II)	Langmuir lead sorption capacity of 14.3 mg/g biochar was reported and significantly reduced its metal toxicity.	90
	Pulp mill sludge	Saturation of zerovalent iron (ZVI) with improved pore structure	Addition of ZVI on the biochar surfaces	Pentachlorophenol	ZVI-MBC and biochar have the capacity to remove the pentachlorophenol at 100% and 73%, after 240 min.	91
Activated biochar	Municipal sewage sludge (MSS)-sugar cane bagasse (SCB		Co-pyrolysis process and activated by using the chemical activation method with the help of phosphoric acid	Pulp and paper industry wastewater	Reduced the COD by 84.61% and color of wastewater by 98.03% at optimized conditions	92
	Activated charcoal -neem leaf powder (AC-NLP)		Activated through carbonization using phosphoric acid	Heavy metals: Cu, Pb, Cr, Ni, Zn and Cd	Highest adsorption capacity was reported by activated biochar for Cu (185.8 mg/g), Pb (205.6 mg/g), Cr (110.9 mg/g), Ni (120.6 mg/g), Zn (133.3 mg/g) and Cd (154.5 mg/g)	93
	Activated neem leaf powder (a-NLP)		carbonization process	Adsorptive batch treatment for individual and combined wastewater	Maximum reduction was achieved up to 82.6−75.9%, 93.4−82.4% and 93.4−86.1% of BOD, COD and color, respectively from wastewater.	94

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reveals the elevation in the randomness of the dye as it has a positive value (0.053 J/mol K). The Freundlich adsorption isotherm best fitted the adsorption isotherm. The pseudo-second-order of the process is shown by kinetic data.[36] Rath et al. (2019) studied the adsorption of Congo red dye using TiO_2 nanoparticles at optimized conditions. They reported that the kinetic analysis, positive values of $\Delta H°$ and $\Delta S°$ indicated significant dye removal. The coefficient of regression (R^2) value determined by Freundlich adsorption isotherm was 0.98m which best fit with the equilibrium data.[37] Table 1 provides a detailed idea about some recent nanotechnological applications for eliminating the heavy metals from industrial wastewater.

The TiO_2, MgO, Fe_2O_3 and aluminum oxides (Al_2O_3) are the most substantially investigated metal-oxide nanoparticles for removing heavy metals from wastewater.[55] However, Engates and Shipley explored the adsorption capacity of TiO_2 nanoparticles for the heavy metal ions, i.e., zinc (Zn), cadmium (Cd), lead (Pb), copper (Cu), and nickel (Ni), from a solution and reported significant adsorption efficiency. The specific surface area of metal-oxide nanoparticles was found to be 185.5. Kinetic data determined the first-order reaction. Langmuir adsorption isotherm was best suited to delineate adsorption. The distribution coefficient (K_d) value revealed that TiO_2 had better adsorption capacity compared to other metal-oxide nanoparticles.[56] Manganese-oxide nanoparticles have better adsorption properties than their bulk counterparts because of their greater specific surface area and polymorphism architectures.[57] Hydrous manganese oxide could be synthesized by adding manganese sulfate monohydrate ($MnSO_4 \cdot H_2O$) to sodium hypochlorite (NaClO) solution, and the Brunauer−Emmett−Teller (BET) surface area calibrated is 100.5 m^2/g.[58] The inner sphere complex is formed by the sorption of the heavy metal ions on hydrous manganese oxides, which could be demonstrated by the ion exchange mechanism.[59] The Freundlich adsorption isotherm is better suited than the Langmuir isotherm, elucidating that the active sites existing on the these oxide surfaces are heterogeneous. Metal adsorption takes place in the order Pb(II) > Cd(II) > Zn(II). Ferric-oxide nanoparticles studied for remediation of wastewater include goethite (α-Fe (OH)O), amorphous hydrous ferric oxides, magnetite (Fe_3O_4) and iron oxides.[55,59−62] The pressure jump relaxation technique was used to determine the kinetic data for Cu(II) adsorption and it is pH dependent phenomenom.[63] It forms an inner sphere complex on the surface of nanogoethite with surface area of 71.49 m^2/g. These nanogoethites reported having a maximum adsorption capacity of Cu(II) with 149.25 mg/g. It follows pseudo-second-order kinetics and adsorption best explained using Freundlich isotherm (R^2 = 0.56−0.57), signifying the homogeneous surface for sorption of Cu(II).[60] Magnetite nanoparticles are prepared by the adding of alkaline carbonate to the Fe(II) and Fe(III) having solution in a molar ratio of 1:2 by precipitation process.[39] The diameter of the magnetite nanoparticles (8.5 ± 1.6 nm) was determined using scanning electron microscopy. For the preparation of composite sorbents, nanomagnetite was used as a magnetic core which helps in the removal of heavy metals.[62,64] Hydrous Fe_2O_3 are prepared by precipitating ammonia with ferric chloride or nitrate in a carbonate-free environment and it has the surface area of 600 m^2/g.[65] The adsorption of Cu(II) and Pb(II) on Fe_2O_3 depends upon the varying concentration of ionic strength from 0.005 to 0.5 M $NaClO_4$.[66] The formation of inner-sphere complexes due to changes in ionic strength may cause a poor adsorption property. The Al_2O_3 nanoparticles are produced by using the sol−gel method and the absorption capacity of these nanoparticles could be enhanced by changing some physical and chemical properties of functional groups.[41,67] The prepared Al_2O_3 was found to be 53 nm in size and used for the sorption of Cd(II), Pb(II), Mn(II) and Cr(III) metal ions, resulting in a good Freundlich adsorption isotherm.[68] In contrast, the Langmuir isotherm was best fitted for Ni(II) and Co(II) ions.[69]

3.1.2. Metal Nanoparticles. Metal nanoparticles are very efficient in wastewater treatment, thus contributing to a safer environment. Various metal nanoparticles, such as zinc nanoparticles, are very efficient in degrading dioxins,[70] silver nanoparticles serve as antimicrobial agents in disinfecting wastewater,[71] iron nanoparticles can remove heavy metal ions,[72] and so on. Iron nanoparticles are coated with Fe and Fe_3O_4. These nanoparticles have a better tendency to absorb some metal ions such as Cu(II), Hg(II), Ni(II), Cd(II), and Cr(II).[72−74] A comparative analysis of Ferric chloride ($FeCl_3$) and iron nanoparticles in removing dark color, COD and BOD concentration was done using 1 g/L $FeCl_3$ and 200 μL iron nanoparticles. The study's results revealed that with 1 g/L $FeCl_3$ concentration, the decrease in color, BOD and COD were 40.76%, 68.88% and 54.16%, respectively, while, after the treatment of industrial effluents with iron nanoparticles, a significant reduction in color (99.10%), BOD (85.92%) and COD (84.16%) was obtained.[12] Therefore, it can be concluded that despite the lower concentration of iron nanoparticles than $FeCl_3$ in the experiment, it was more effective than $FeCl_3$ in reducing color and BOD and COD concentration. Iron nanoparticles tend to form free iron ions in the vicinity of oxygen and water, in addition to their large surface area; hence, they are highly reactive in nature.

3.2. Use of Nanocomposites. Nanocomposites such as biochar and chitosan/nanocellulose membranes, remove dyes and heavy metals from contaminated synthetic wastewater, respectively. Adsorbents such as biochar based nanocomposites, are produced from pulp and paper sludge. At the same time, the composite membrane is formed from chitosan and cellulose, which are naturally occurring polysaccharides and are biodegradable.[75]

3.2.1. Biochar Based Nanocomposites. Biowastes are either burnt or disposed of as debris, adversely impacting the environment and public health. An economical approach toward converting biowastes into material containing carbon, such as biochar, could pave the way to remove dye substances within a shorter period than raw biomass and would be a cost-effective method. Biochar based nanocomposites are derived from pulp and paper sludge and help in the removing the dye from synthetic wastewater.[76] The synthesis, characterization, and performance of biochar based nanocomposite materials are given in Table 2. A study was conducted to remove methylene blue dye from synthetic wastewater using biochar and Fe_2O_3 biochar nanocomposite.[77] This dye has been used as a coloring agent in paper, hair, and textile dyeing. At lower concentration, it does not cause precarious effects but at higher concentration it can lead to severe health issues concerning mental disorders, hypertension, jaundice, and fever.[78,79] Adsorbents were prepared by pulp and paper sludge, which undergoes further processing. The sludge was dewatered and then air-dried for 2 days before being dried in the oven for 24 h at 70 °C. The first half of the dried part was enclosed in a steel container to

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produce biochar, and another part was soaked in $FeCl_3 \cdot 6H_2O$ solution. Characterization of biochar and Fe_2O_3 nanoparticles showed that they have heterogeneous surfaces and pore sizes ranging from 1.7 to 300 nm.

Hence, they furnished good adsorption properties for methylene blue dye at pH 12−14. In comparison, it was concluded that the maximum adsorption capacity of the Fe_2O_3-biochar nanocomposite (50 mg/g) was relatively higher than that of pristine biochar (33 mg/g). This is due to adsorption on surfaces such as Fe_2O_3 nanoparticles and the biochar matrix. Freundlich and Langmuir's isotherms gave a close estimation of adsorption data for biochar and Fe_2O_3-biochar nanocomposite.[95] Although they have less adsorption capacity than any other adsorbent, their utility in wastewater treatment is an innovative approach to improving environmental conditions and reducing health risks.

3.2.2. Chitosan-Nanocellulose Based Composites. Metallurgical industries, mining operations, tanneries, etc., release harmful effluents consisting of metal ions that are toxic to water bodies and thus have an adverse effect on the environment and public health. The metal ions released from these industries are considered heavy metals because they are toxic in lower concentrations.[96] Recently, nanoscience and nanotechnology have emerging solutions to cope with industrial effluents. However, nanostructures having physical and chemical properties have demonstrated significant degradation of effluents.[97,98] The nanocellulose based adsorbent has excellent merits including high specific surface area, stable in water (reduce biofouling) and its ability to be surface modification or functionalization for a wider applications.[99] A study was performed on removing chromium ions from wastewater using chitosan/nanocellulose based composites.[75] The delignification and acid hydrolysis method were used to extract nanocellulose from sugar cane bagasse, and these nanocelluloses could be used as a source material for the preparation of sustainable composite material. A nanocomposite membrane was formed using a solution casting method and examined to determine their ability to remove chromium ions using vacuum filtration units. Chromium ion concentration between 87 and 29 ppm was removed efficiently within the first four cycles.[100] The Fourier transform infrared (FTIR) and X-ray diffraction (XRD) techniques were used for the characterization of the resulting membrane. An analytic study exhibited the crystalline order of the membrane using XRD and a cellulose structure with characteristic peaks. The composite membrane is hydrophilic, having a contact angle of 60.7°. The maximal swelling ratio achieved is 1.8 in 5 min. Earlier research on swelling capacity indicated that if the amount of nanocellulose is elevated on the chitosan matrix, then the swelling capacity of the composite membrane is reduced; i.e., the membrane becomes less permeable.[101−103] Results of chemical stability determined that the membrane dissolves entirely in the acidic solution, whereas it swells in the primary solution. Composite membranes have immense potential compared with conventional methods to remove industrial effluents comprising heavy metal ions. Moreover, it is also significant in removing harmful dyes and other toxic substances. Electrosterically stabilized nanocrystalline cellulose has been developed by Tavakolian et al. and stated that the adsorption of 1400 mg methylene blue dye per gram of adsorbent.[98] The prepared porous composite material in combination with limestone and chitosan based cellulose has adsorbed 130.75 mg/g of Cu^{2+} and showed pseudo-second-order kinetics by satisfied Freundlich model during the removal process.[104] Another study conducted by Bambaeero and Bazargan-Lari and reported the chitosan and hydroxyapatite obtained from snail and prepared the snail shell/hydroxyapatite/chitosan composite for removal of Cu^{2+} and Zn^{2+} ions from solution by a removing efficiency of 60% for Zn^{2+} and 90% for Cu^{2+} ions.[105]

3.3. Use of Nanoadsorbents. Nanoadsorbents such as graphene and carbon nanotubes were used for the elimination of heavy metal ions and phenolic compounds from synthetic wastewater and waste materials such as agro residues, municipal waste, food processing waste, etc., respectively.[106,107]

3.3.1. Carbon Nanotube Adsorbents. Carbon nanotubes are carbon based materials used for remediation of aqueous solutions consisting of heavy metal ions.[72,108,109] Adsorption using carbonaceous materials is a potential and economical way to remove heavy metal ions.[110] Carbon nanotubes are hollow and cylindrical and are formed in a rolling pattern from single or multiple graphene sheets.[111] Characteristics include large surface area, hydrophobic membrane and easy modification.[112] They have magnificent electrical, thermal, and mechanical features.[113] These adsorbents possess different functional groups. Physical or chemical interactions between functional groups of adsorbents and heavy metals are crucial for the adsorption of heavy metals.[114] Functional groups are classified into oxygen-, nitrogen-, and sulfur-containing functional groups based on the bonding of heteroatoms on the carbon surfaces. Modification of carbon surfaces results in enhancement of their ability to selectively adsorb heavy metals.[115,116]

3.3.2. Graphene Nanoadsorbents. Graphene nanoadsorbents have an immense potential to reduce environmental pollution because they have a high surface area, $\pi-\pi$ bonding, and chemical longevity and can be produced on a wide scale. Graphene is a two-dimensional structure packed tightly in a honeycomb-like crystal lattice.[100] The graphene oxide (GO) having surface area of 2620 m^2/g is used for the wastewater treatment.[117,118] A study was demonstrated for the removal of phenolic compounds from the discharge of industries, including refining, resin production, pulp and paper, and pharmaceuticals, and found that graphene nanoparticles have great potential to remove phenolic compounds.[106] Similarly, chitosan reinforced graphene oxide-hydroxyapatite (CS@GO-Hap) based composite matrix was utilized for the elimination of the organic contaminants and it showed significant adsorption efficiency by adsorption of Congo Red (43.06 mg/g), Acid Red 1 (41.32 mg/g) and Reactive Red 2 (40.03 mg/g) dyes from effluent water, respectively.[119] A similar study conducted by Nguyen et al. using the composite materials like hydroxyapatite/graphene oxide/chitosan beads (HGC) for the sorption of heavy metal ions and dyes and reported the notable sorption capabilities of the HGC beads were 256.41 mg/g for copper ions and 99.00 mg/g for MB dye.[120] Banana peel-reduced GO (BRGO) was explored for the elimination of the aqueous Cr(VI) and acid dye, resulted maximum adsorption capacity for Cr(VI) (135.87 mg/g) and AV54 dye (110.74 mg/g).[121]

4. OTHER ADVANCED METHODS FOR TREATMENT OF PULP AND PAPER WASTEWATER

4.1. Electrocoagulation Based Methods. Electrocoagulation is an emerging technological area that can eliminate different types of contaminants present in effluents.[122] It is a

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Table 3. Some Advanced Nanotechnological Applications for Mitigation of Pollutants and Their Significance of Study

Type of Nanomaterial	Pollutant type	Adsorption/Removal/Sorption efficiency	Significance of study	Reference
Nanoparticles				
Zinc-oxide nanoparticle	Organic pollutants from the paper mill effluent sample	86%	ZnO NPs were shown to be highly effective at eliminating organic pollutants and to have a multiple reuse capacity.	126
Zinc-oxide nanoparticle	Congo red	48.3 mg/g	Removed 92% and 87% of the congo red and malachite green, respectively	127
	Malachite green	169.5 mg/g		
Cobalt and cobalt-oxide nanoparticles	Murexide dye	43.6% using Co-nanoparticles (chemically synthesized)	For the purpose of remediating wastewater containing dyes, the microwave synthesized method should be used to prepare NPs because it is quick, affordable, and environmentally friendly and produced catalysts with a better comparative photocatalytic degradation efficiency toward murexide and EBT dyes than the chemical method.	128
		39.4% using Co_3O_4 nanoparticles (microwave synthesized)		
Steel waste based iron-oxide nanoparticles	Lead(II) and chromium (VI) metal ions present in synthetic water and industrial wastewater	up to 99.9%	Successful removal of chromium and lead contaminants from wastewater was observed.	129
Iron nanoparticles - *Azadiracta indica* extract iron nanoparticle	Phosphate	98.08%	The study showed great promise for an in situ treatment method capable of removing COD, phosphate, and ammonia all at once.	130
	Ammonia	84.32%		
	Nitrogen	82.35% of chemical oxygen demand		
	Chemical oxygen demand from domestic wastewater			
Fe_2O_3 nanoparticles	Methylene blue photocatalytic remediation under visible light irradiation	97%	Remarkable capacity of Fe_2O_3 nanoparticles to degrade methylene blue was observed. Also, F_e2O_3 nanoparticles displayed exceptional stability.	131
Nanocomposites				
Biochar based nanocomposites	Chromium(VI)	102.66 mg/g	Synthesized nanozinc particles combined with biochar presented new opportunities for the economical and successful removal of Cr(VI) and other heavy metal pollutants from wastewater.	132
Chitosan-nanocellulose based composites	Chromium ions	87%	The heavy metals removal from composite membranes based on chitosan and nanocellulose demonstrated great potential.	75
Titanium doped activated carbon−cellulose nanocomposite	Organic pollutants - crystal violet and methyl violet	Nearly 87%	The ability of the evaluated nanocomposite to remove dye has primarily been attributed to surface adsorption, with the help of photocatalysis.	23
Nanoadsorbents				
Carbon nanotubes adsorbents: Multiwalled carbon nanotubes and carboxylated multiwalled carbon nanotubes	Arsenic(V) and Manganese (VII)	200 and 192 mg/g for multiwalled carbon nanotubes, respectively	The research demonstrated that carboxylation of carbon nanotubes improved manganese (VII) and arsenic(V) adsorption.	133
		250 and 298 mg/g for carboxylated multiwalled carbon nanotubes, respectively		
Graphene nanoadsorbents: Chitosan/graphene oxide	Gold(III)	80−500 mg/L	Chitosan combined with 5 wt % graphene oxide, can improve Au(III) and Pd(II) adsorption capability and be effectively utilized for Au(III) and Pd(II) recovery throughout a wider pH range.	134
Porous graphene nanoadsorbent	Methylene blue	99.69%	APG significantly encourages the decrease of organic (COD) and solid contents (TDS) in addition to dye removal.	135
Sulfonated magnetic graphene-oxide composite (SMGO)	Copper(II)	73.71 mg/L	The adsorption reaction of Cu(II) on the SMGO was shown by the thermodynamic parameters to be an endothermic and spontaneous process. These findings suggest that the SMGO is an excellent material for cleaning up metal ion pollution.	136
Functionalized graphene oxide (FGO) - Graphene-oxide modified waste newspaper	Pb^{2+}	75.41 mg/g	GO constructed paper absorbents found to be effective and affordable adsorbent that may be used in the pulping and papermaking industries since they were easily removed from the solution.	137
	Ni^{2+}	29.04 mg/g		
	Cd^{2+}	31.35 mg/g		

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Table 3. continued

Type of Nanomaterial		Pollutant type	Adsorption/Removal/Sorption efficiency	Significance of study	Reference
Advanced Methods					
Electrocoagulation based methods		Resin acid and copper	97%	A possible technique for removing harmful contaminants from pulp mill wastewaters was applied by combine electrocoagulation with separation methods like filtration, sedimentation, or flotation.	4
Photocatalysis based advanced oxidation methods	ZnO photocatalyst	Genotoxic azo dye Acid Violet 7	Complete degradation occurred in the presence of ZnO and UV light at 60 min irradiation while in the case of TiO_2-P25 under same conditions, only 57.5% degradation occurred.	ZnO was found to be more effective at degrading AV 7 than TiO_2-P25.	138
	Fe−TiO_2 composite	Reduction of color from pulp and paper industries secondary treated wastewater	91.6%	Using waste materials to create the catalyst support turned out to be an innovative approach for handling the P&P industry's color-permanent effluent.	139
	Jumbo muscadine extract based cobalt-oxide nanoparticles	Acid Blue-74 dye	98%	Excellent photocatalytic activity was demonstrated by the synthesized GCoO-NPs, and in the future, the NPs' essential biological components may be used to treat microbes.	140
Adsorption by carbon and polymer resins	Activated carbon and polymer resin-polystyrene divinylbenzene co-polymer	Color removal of wastewater	95%	Polymer resin showed great promise as an adsorbent for removing color from bleach plant effluents.	141
Integrated process	Permanganate, electro-Fenton and Co_3O_4/Peroxymonosulfate/UV (sulfate radical)	COD reduction	Reduced from 1450 to 62 mg/L	It was found that the combination of permanganate, electro-Fenton, and Co_3O_4/Peroxymonosulfate/UV (sulfate radical) was successfully effective in removing COD.	142
	Coagulation-flocculation and solar photocatalysis−based advanced oxidation process using TiO_2-reduced graphene-oxide nanocomposite	COD reduction	95%	The research proved the effectiveness of treating actual pulp and paper mill effluent with a combination of flocculation and coagulation, succeeded by solar photocatalysis utilizing TRGO nanocomposite.	143

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method that involves electrodissolution soluble anodes to form a floc of metal hydroxides inside the effluents to be treated.[123] Electrocoagulation has various advantages, such as being easily operatable, small particles having more affinity for coagulating than bigger ones and sludge being produced in lower proportion.[124,125] A study was showed by Uğurlu et al.[123] aiming to remove lignin and phenolic compounds using electrocoagulation from paper mill effluents. Analysis was performed at 21−22 °C and 200 rpm agitation rate. Observations were recorded at varying current intensities by iron and aluminum electrodes at varied electrolysis time intervals, i.e., 1.0, 2.5, 5.0, and 7.5 min, respectively. Parameters used to eliminate these effluents were 2 min electrolysis time, 77.13 mA current intensity, and 12 V voltage applied. Relatively, aluminum electrodes have better efficacy than iron, having removal capacities of 80% of lignin and 98% of phenol. Removal is enhanced by increasing current intensities.[123] Vepsäläinen and co-workers conducted a study regarding the disposal of toxic effluents in pulp mills by the electrocoagulation method. Wood resin and metals, such as copper or pure resin acids, were present in wastewater that underwent treatment. This technique was able to remove resin acids very efficiently. At pH 5 and a concentration of 125 mg/L, 97% removal was observed when the sample was treated for 60 s. High efficacy in reduction was achieved in pimaric-type acids rather than abietic-type. 72−97% of copper was eliminated while applying 2 A current on the sample solution for 60 s.[4] Electrocoagulation does not have a profound effect on the bacterial toxicity. The EC_{50} value for copper was calculated to be 17.0 mg/L, and for wood resin, it was 38.2 mg/L in *Vibrio fischeri* bacteria. In response to *Raphidocelis subcapitata*, formerly known as *Pseudokirchneriella subcapitata* (microalga), toxicity was removed entirely within a 60 s treatment period. The impact of initial pH, current applied, and period of treatment of the sample on effluent removal was investigated using statistical experimental design and partial least-squares modeling. Electrocoagulation aided in the elimination of color. With an intermediary current flow, a prolonged treatment duration, and relatively high pH, the model predicts optimal elimination. A considerable amount of DOC was also eliminated with protracted treatment time and high current. Electrocoagulation, in combination with separation techniques like filtration, sedimentation, or flotation, could be employed to eliminate harmful pollutants from wastewater released from pulp mills. Ferrous ion oxidation to ferric ion is improved by adding oxidants to the wastewater, which enhances the electrocoagulation.[4] Some literature surveys based on nanotechnological application in effluents treatment and their significance of study are summarized in Table 3.

4.2. Photocatalysis Based Advanced Oxidation Methods. Photocatalysis is an advanced oxidation method regarded as a powerful wastewater treatment technology with various applications in depleting environmental contaminants. This method oxidizes hazardous substances without producing secondary wastes.[144] Characteristic features such as surface area and particle size of the catalyst are essential to understand as reactions in photocatalysis occur on the surface of semiconductors.[145] Nanomaterials (NMs) are used as semiconductors if particle size decreases; specific surface area will increase, increasing the adsorption ability. In photocatalysis, higher activity is observed on the surface.[146,147] Arsenic(III) is completely oxidized to arsenic(V) and eliminated from the drinking water by adsorption from TiO_2 solution.[148] Fenton-like reaction effectively eliminated both the organic pollutants in the presence of hydrogen peroxide without requiring the use of UV light.[149] In the experiment conducted by Nogueira et al., it was found that nano-TiO_2 has higher efficiency than nano-Fe_2O_3 because of its high surface-to-volume ratio. Nano-TiO_2 (<25 nm) nanoparticles are smaller in size than nano-Fe_2O_3 (84 × 425 nm). The surface area of nano-TiO_2 ranges between 45 and 55 m^2/g, and for nano-Fe_2O_3 it is 50−70 m^2/g. The remarkable reduction was seen by the TiO_2/H_2O_2/UV system in the elimination of color (93.3%), aromatic compounds (54.6%) and COD (89.8%) of the pulp mill wastewater at pH 3.0, 0.75 g/L nano-TiO_2 concentration, 2 h of exposure to UV light and 75 mM H_2O_2 concentration. Increasing H_2O_2 concentration in Fe_2O_2/H_2O_2/UV system eliminates the toxicity of pulp mill effluents to *Vibrio fischeri*.[150]

4.3. Adsorption by Carbon and Polymer Resins. After the bleaching process, effluents released from the pulp industries contain lignin and some organic and inorganic compounds that are toxic to the environment. These pollutants form unpleasant odor, color, and foaming.[151] These compounds are recalcitrant and cannot be easily degraded by conventional methods, such as coagulation, sedimentation, and filtration. However, the adsorption technique is paving a new way in the treatment of these effluents; hence, the used adsorbent must be regenerated, keeping the cost of regeneration.[152] Polymer resins have advantages over activated carbon as they could be restored by processing with acid and alkali solutions at standard conditions.[153] The resin was more efficient for color removal than the activated carbon.[154] Zhang and Chuang studied the adsorption of organic effluents released after the bleaching process from a pulp mill on activated carbon and polymer resins (polystyrene divinylbenzene copolymer). Moreover, total organic carbon and the efficiency of color removal were also determined by the study. The adsorbent is added to wastewater and kept in a mechanical shaker for 72 h before taking final readings. BET surface area of activated carbon and polymer resin was 723 and 33 m^2/g, respectively, and pore size ranging between 5 and 10 nm indicated the pore volume of activated carbon and polymer resin to be 0.0420 and 0.0587 cm^3/g, respectively. The Freundlich adsorption isotherm at pH 1.96 provided the R^2 value of activated carbon to be 0.99 and polymer resin to be 0.96. The significant adsorption of organic carbon was observed by the polymer resin and activated carbon. Loaded polymer resin can be regenerated by washing with 3 N sodium hydroxide solution.[141]

5. ECONOMIC ASPECTS AND ENVIRONMENTAL SUSTAINABILITY

The use of nanotechnology for the remediation of industrial pollution has significant promise for providing a clean and eco-friendly environment. With the expansion of sustainable nanotechnological applications in wastewater treatment, certain limitations minimize the impact of this technology on effluent treatment. For instance, cost-effectiveness, reusability, and potential environmental impact might be significant factors for the sustainable implementation of nanotechnology in the industrial sector. At the industrial level, the employment of these nanotechnologies is majorly dependent upon their best performance and affordability.[155] In this scenario, a literature survey is regularly reviewed to identify innovations in preparing low-cost sorbents for contaminants. This practice also provides an avenue toward problem-solving the weakness

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encountered using such NMs.[156,157] Another important factor, such as public acceptance, is one of the valuable criteria for pushing nanotechnology for commercialization. The commercialization of academic research offers a mechanism to directly impact the economy and society and serves as a gauge of scientific productivity. Although research on wastewater treatment is currently dominated by nanotechnology, there are no reliable studies regarding any current commercial multipollutant control nanotechnologies. However, patents might clearly indicate a technology's potential for commercialization.[158] Thus, it would be necessity to deliberate the social benefits rather than technical and economical values while developing the sustainable technology for wastewater treatment plants.[159,160] According to the literature, in the present scenario, there are so many gaps in knowledge on the applicability of nanotechnology in wastewater treatment.[161−164]

All types of NMs showed excellent specific areas and promising capacities for removing effluents from the environment. In contrast, some issues must be resolved to improve their qualities and make them more appropriate for wastewater applications.[165] Moreover, NMs have significant disadvantages, such as agglomeration and aggregation in the wastewater, so they are reinforced using some reinforcements. The aggregation of these NMs causes several toxicity and health issues. Hence, scientists have explored addressing these issues by introducing them into several supporting and bulk sorbents. Hence, nanocomposites material is one of them, which act as support and enhance the surface properties of the nanomaterials.[166] Despite the advantages of NMs, their toxicity, bioavailability, long-term environmental persistence, and bioaccumulation must be considered. Due to their diminutive size, they can adsorb environmental contaminants and bind with organic compounds to form toxic substances. The formed toxic compounds moved along with the pollutants and might cause secondary pollution. This phenomenon poses a significant challenge to the researchers. To overcome these problems, immobilizing the NMs in a matrix may reduce the release of hazardous toxic compounds in the aquatic water system.[167,168] Finally,[169] the nanotoxicity of NMs in the environment must be considered. However, studies have reported that the size-dependent properties of NPs must be regarded as nanotoxicities in the environment. Legalizing the regulatory bodies to scrutinize the applicability of NMs in the biotechnological process and release into the environment would be necessary. The size and chemical properties of these NMs must be checked in terms of the in vitro toxicity. By addressing the above limitations, NMs will demonstrate admirable materials for the treatment of industrial wastes in the near future.

6. CONCLUSION AND FUTURE PROSPECTIVE

Challenges in global pollution resulting from rapid urbanization and industrialization require innovative treatment technologies to overcome environmental issues. Recent scientific research has led to the recognition of nanotechnology applications for environmental sustainability and preservation. Nanomaterials offer several essential physicochemical topographies that brand them particularly appealing for wastewater treatment. It has a great affinity to toxic effluents of organic and inorganic compounds. These nanomaterials can be functionalized with different chemical groups to combat the conventional/traditional treatment process and have excellent treatment capability. Recent knowledge about employing NMs to mitigate pollutants from wastewater through adsorption and photocatalysis is described in a well-structured article having challenges that are presently faced in the scenario. The present review concluded that the innovative technologies have significant effluent treatment ability and could be employed to treat industrial wastewater containing dyes and heavy metals along with other hazardous organic compounds. Overall, carbon based materials with designed functional groups may be promising adsorbents for different environmental biotechnological applications.

Furthermore, there are various important factors that need further research under active investigation and future research based on the application of NMs in wastewater treatment. Therefore, some few directions need to be explored: (i) The current nanomaterials often require strong buffer capability, robust treatment, and intensive physicochemical reactions. (ii) It is necessary to produce novel, facile, cost-effective, and environmentally friendly technologies to mitigate the environmental load. (iii) More research is needed in the comparative study between recyclability and regeneration of NMs in the treatment systems at scale up level. (iv) Many other pollutants coexist that strongly influence the degradation and removal efficiency. Hence, more investigations and strategies are required to understand the causal mechanisms that govern the inhibitory interactions.

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Notes

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