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Functionalization of Carbon-Based Nanostructures for Environmental Remediation

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Abstract

The rise of environmental exploitation due to human intervention and industrialization is an alarming issue at current times. Carbon-based nanostructures have been in the application of environmental remediation due to their high adsorption nature, low toxicity, and other beneficial properties. Functionalization of the carbon nanostructures with relevant functional groups improves the performance of the material, which results in the target-specific application. This review focuses on the role of functionalization of carbon-based nanostructures for environmental remediation purposes. Methods of functionalization, properties, and the applications of functionalized carbon nanostructures are also discussed.

1. Introduction

The cause of water scarcity is not always due to the lack of water but the contamination of already available water resources. Although water is the most indispensable substance for the survival of life on Earth, it is constantly being deteriorated. The main sources of water pollution are organic dyes, pharmaceuticals, cosmetics, pesticides and heavy metals released by human intervention and industrialization [1]. This makes the water bodies unsuitable for regular use. Besides being the primary source of pollutants, industrial effluents also cause adverse health effects on human and aquatic environments. Remediation using pertinent materials is an urgent need of an hour to deal with this problem [2-4]. A wide range of conventional methods of water treatment includes extraction, isolation, filtration, and ion-exchange. However, adsorption and photocatalysis using nanomaterials is the most approachable technique as it is simple, budget-friendly, highly efficient, and cost-effective comparatively [5]. Nanomaterials are chosen over bulk materials because of their increased surface area and emergence of unique size-related properties. Nanomaterials of metal, metal-oxides and carbon-based materials have been in the action of environmental remediation over recent years [6]. In the past decade, carbon-based nanostructures are highly preferred for applications in water remediation due to their high adsorption properties and low toxicity. In recent studies, incompatibility, poor dispersion, low surface activity and other drawbacks of carbon nanomaterials have been overcome by functionalization of the material with appropriate agents. Functionalization can be carried out by either physical or chemical methods. Physical methods involve coating or wrapping the material through physical forces of attraction, whereas chemical methods involve the reaction of pristine carbon nanomaterial with functionalizing agents and the formation of chemical bonds. Functionalized carbon nanomaterials have been successfully employed for water treatment of heavy metals such as cadmium, chromium, arsenic, lead, mercury, and dyes such as methylene blue and Congo dye through adsorption and photocatalytic methods [6-10]. This article provides an overview of sustainable synthetic approaches to carbon nanostructures and functionalization of carbon nanostructures specifically with nitrogen and iron functionalization are used for environmental remediation applications. Apart from nano adsorbents for water remediation, carbon-based nanostructures are also employed for monitoring of toxicants in soil and food, electrochemical sensors, gas sensors, conversion of toxicants to into useful products, electrodes for energy storage batteries etc.

2. Nanomaterials in the Action of Remediation

Nanomaterials are materials with any structures in nanoscale dimensions. Nanostructures can be spherical, tubular, cylindrical, or flower-shaped etc. Due to the increase in surface area as size decreases, nanomaterials possess beneficial properties. One of the main applications of nanomaterials in environmental remediation is removal of toxic metals and dyes because of their high porosity, large surface area and optical properties such as bandgap. Several nanomaterials of transition metals, metals and metal-oxides are employed for remediation and mitigation purposes. Many nano adsorbents, photocatalytic nanomaterials, magnetic nanomaterials, nanoscale zero-valent materials etc., have been in application for water remediation [11]. Carbon dots, carbon nanotubes (CNTs), carbon aerogels, graphene, graphene oxide, fullerenes have also been successfully employed for water remediation concerning heavy metal treatment, removal of industrial eluents and other contaminants [8]. Many metal-based nanomaterials are highly used in water treatment techniques; however, their fate and post-treatment toxicity is of high concern [12-13]. This opens an avenue for carbon-based nanomaterials, while functionalization of carbon-based nanomaterials imparts less toxicity and higher efficiency.

3. Carbon Nanostructures

Carbon, the element of life, is one of the most abundant non-metals present on Earth. Carbon is most popular for its wide range of allotropes with unique properties. Carbon nanostructures are highly encouraged because of their unique properties at the nanoscale [14]. While the carbon bulk forms the basis of life, the carbon nanostructure promotes life on Earth. Carbon nanostructures find application in various sectors such as energy storage, sensors, biomedical and environmental remediation due to their exciting properties. Carbon nanostructures, due to their less-toxic nature, are widely used for environmental remediation purposes [15]. Carbon nano-allotropes can be classified based on their dimensions. For example, graphite and carbon aerogels are three-dimensional, graphene derivatives are two-dimensional, carbon nanotubes are one-dimensional and carbon quantum dots are zero-dimensional [16]. The synthesis, properties, environment and energy related applications of the various carbon nanostructures are discussed here.

3.1 Graphene Derivatives

Graphene is the basis of all other carbon nanoallotropes. Graphene is a single layer of graphite and a two-dimensional planar sheet of one atom thickness consisting of sp2 hybridized carbon atoms with in-plane σ bonds and out-of-plane ⍰ bonds. Graphene is the thinnest and softest material known today and shows excellent properties such as high thermal and electrical conductivity, improved optical transparency, mechanical stiffness, and high surface area [17]. Many synthetic methods of graphene such as mechanical exfoliation of graphite, chemical vapour deposition, epitaxial growth on SiC, electrochemical exfoliation and carbon nanotube unzipping etc are known [18]. As graphene is expensive and hard to synthesize, graphene derivatives such as graphite oxide, graphene oxide and reduced graphene oxide are preferred. Graphite oxide is formed by Hummer's method where graphite on reaction with sulfuric acid creates exfoliation and intercalation of graphite flakes. Graphene oxide is formed on addition of strong oxidizing agents such as $KMnO_4$ to graphite oxide, leading to introduction of oxygen functional groups [19]. This differentiates graphene oxide from graphite oxide structurally. Graphene oxide with more oxygen functional groups on its surface is strongly hydrophilic while reduction of oxygen groups produces hydrophobic reduced graphene oxide. Reduced graphene oxide is synthesized from graphene oxide chemically using suitable reducing agents. It can also be reduced thermally by heating at a higher temperature in the microwave. Electrochemical reduction can be an alternative method as it is non-toxic and easy and prevents damage to material at high temperatures [20]. Many functionalized graphene-based nanocomposites have shown outstanding performance for environment and energy applications [21].

3.2 Carbon Aerogels

Carbon aerogels are ultrafine porous structures with low density, large specific surface area, pore volume and exceptional adsorption and conductivity. It was initially obtained upon sol-gel polycondensation of formaldehyde and resorcinol by pyrolysis at high temperatures. Recently, carbon aerogels are derived from natural polymers and biomass wastes as a sustainable approach. Due to their excellent mechanical strength and environmentally stable properties, they are highly employed for environmental remediation techniques [22].

3.3 Carbon Quantum Dots

The fascinating class of carbon nanoparticles are carbon quantum dots (CQDs), which are composed of carbon atoms of diameters of about 10nm. CQDs are fluorescent nanomaterials with highly controllable photo-luminescence and optoelectronic capabilities due to their potent quantum confinement effect. The oxygenated functional groups have changed the surface structures and particle sizes of CQDs, resulting in the display of quantum confinement effects. CQDs can be prepared by various methods such as ablation, pyrolysis, solvothermal, hydrothermal and microwave methods. Typically, CQDs possess hydrophobic sp^2 hybrid carbon cores covered by hydrophilic oxygen and nitrogen functional groups on the surface, resulting in stable aqueous dispersion and biocompatibility [23].

3.4 Carbon Nanotubes

Carbon nanotubes were first produced by Iijima in 1991 [24]. CNTs are nothing but graphene sheets rolled up to a tubular structure forming a one-dimensional carbon structure with considerable length, size, and chirality. Carbon nanotubes are generally synthesized by laser ablation, arc-discharge methods, chemical vapor deposition, electrolysis, and hydrothermal methods [25,26]. Carbon atoms in the CNTs are sp^2 hybridized. Based on the wall's structure, the carbon nanotubes are classified into single-walled CNTs, double-walled CNTs (composed of two cylindrical tubes) and multi-walled CNTs (composed of several concentric tubes). The graphene nanosheet is rolled along a (m, n) lattice vector into tubular SWCNTs, where (m, n) refers to the chiral indices. Mechanical, optical, and electronic properties depend on the value of the lattice vector [27]. SWCNTs can be classified based on the chiral vectors as zigzag (m= 0), armchair (m= n) and chiral ($m \neq n$), for example see Fig. 1. In the case of multi-walled CNTs, each layer of graphene can have a different chirality. The chirality of the nanotubes notably influences their properties. Carbon nanotubes possess unusual mechanical properties with high Young's modulus and tensile strength, physical properties such as high thermal stability and electrical properties [28]. CNTs have been reported to be toxic because of their tubular structure. Functionalization reduces toxicity. However, a sustainable approach of functionalization is recommended [29].

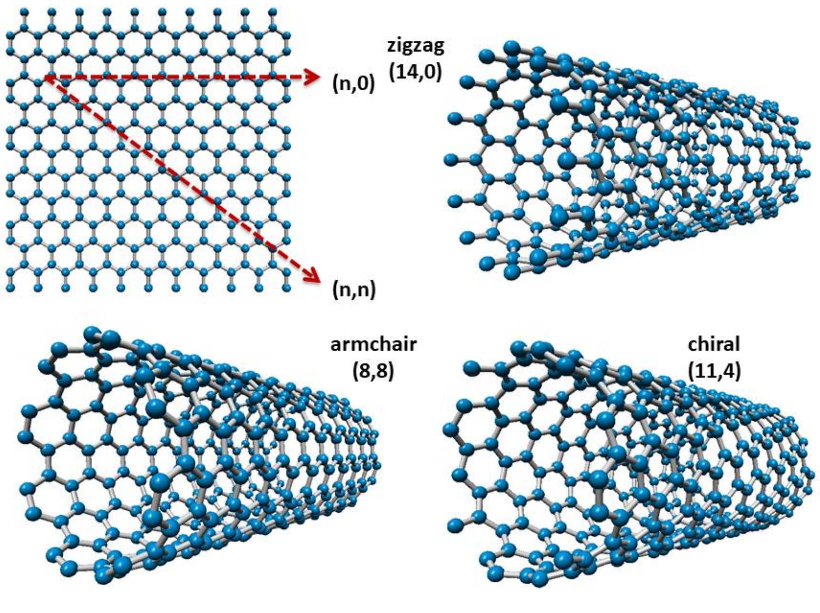

Fig. 1 *Types of carbon nanotubes based on chiral vectors*

Apart from carbon nanotubes, graphene and graphene oxides, other carbon-based nanostructures have been known for environmental remediation purposes. Carbon-nitrogen materials are known for their unique electronic and catalytic properties. Typically, the highly stable graphitic carbon nitride with a planar π-conjugated structure is employed for the removal of water ions from wastewater [30]. Nanoscale carbon flakes, carbon fibers, aerogels and carbon dots were also used for removal and detection of metal ions and other pollutants in drinking water and wastewater. One-dimensional carbon nanofibers and two-dimensional carbon nanoflakes are made of strong covalent C-C bonds, and comparatively less expensive compared to CNTs [14,31]. They exhibit excellent performance in adsorption and as anode material in energy storage. Activated charcoal, a highly porous carbonaceous structure produced from green precursors has been widely used as adsorbents in water treatment because of its high porosity and large surface area [32].

4. Functionalized Carbon-Based Nanostructures

The concept of functionalization was introduced to solve issues of non-functionalized carbon-based nanostructures concerning dispersibility, compatibility and surface reactivity etc. In simple terms, functionalization leads to surface modification of the material.

Functionalization can be of two types, namely, covalent, and non-covalent functionalization. Covalent functionalization is attained through covalent bond formation while non-covalent functionalization is attained through Van der Waals forces. Functionalization can be done by physical or chemical methods. The method by which carbon nanostructures are bonded to the functional groups via chemical reactions is called chemical functionalization. Whereas methods such as wrapping and coating of carbon nanostructures using functional groups, encapsulation of functional groups in carbon nanostructures is called physical functionalization. Various methods of functionalization include chemical reduction, chemical grafting, in-situ polymerization, hydrothermal, pyrolysis, sonication, amination etc. [33]

4.1 Synthetic Approaches

There are many methods for the synthesis of functionalized carbon nanostructures, namely, acid treatment, chemical treatment, cross-linking, hydrothermal methods, pyrolysis, and electrodeposition etc [34]. The sustainable approach often uses low-cost natural precursors, waste materials and biomass to synthesize useful functionalized carbon nanostructures. The one-step method, one-pot synthesis, simultaneous and in-situ functionalization is a direct or facile way of approach. The choice of method depends on the aim of functionalization and oriented based on their application.

4.1.1 Natural Precursors and Biowaste

Biowastes are transformed into useful materials by this green method of approach. For instance, two-dimensional carbon flakes have been synthesized from waste onion sheathing (*Allium cepa*), which allowed facile functionalization of the surface with magnetic Fe_3O_4 nanoparticles for application in arsenite adsorption. *Allium cepa* has been a good choice of green source because of its abundance as waste material and its unique two-dimensional shell structure [35] (Table 1). Similarly, waste eggshells have been used as both reactor and bio template for the synthesis of carbon fibers coated with Co_9S_8 nanorods by in-situ carbonization and sulfurization. Eggshells play a key role as the collagen; a protein fiber and dermatan sulphate, chondroitin tends to be the carbon and sulphur source respectively. Intrinsic protein fibers are carbonized into carbon fibers while sulfur content was employed to transform $Co(OH)_2$ to Co_9S_8 nanorods [36].

Wastewater stream from the olive oil mill industry, which is highly phytotoxic, was used as a rich carbon source for the synthesis of thin shell carbon encapsulated iron nanoparticles through hydrothermal carbonization [37]. Ordered mesoporous magnetic carbon (OMMC) was synthesized from a natural precursor, curcumin. OMMC along with magnetic ferric nitrate nanoparticles is used as an adsorbent in solid-phase microextraction (SPME) of gadolinium from wastewater samples [38]. Biomass waste such as chitin, cellulose and lignin are widely considered as a source of carbon-rich precursor materials. Porous carbon was fabricated from a biomass waste sorghum on which Pt and Pd nanoparticles are then confined by a supercritical CO2 deposition method. This novel functional material helps in the selective conversion of biomass-based furfural into furfuryl alcohol and tetrahydrofurfuryl alcohol [39].

Lotus leaves were used as a biomass carbon source that on pyrolysis along with KOH produced diverse carbon nanostructures [40]. Waste biomass such as vine shoots and eucalyptus trunk wood have been used as a carbon source for the preparation of sodium vanadate fluorophosphate functionalized carbon material via hydrothermal carbonization, which finds application as the cathode in Na/Li-ion battery due to its improved electrochemical performance [41].

Fluorescent N, S, P co-doped carbon nanodots have been synthesized by one-step hydrothermal pyrolysis from Saccharomyces cerevisiae as a waste microorganism precursor. The doped carbon material showed high stability and sensitivity towards the detection of manganese ions and L-Ascorbic acid in biosystems compared to classical probes [42]. Seafood waste rich in chitosan has been used as a source of nitrogen for nitrogen doping, resulting in enhanced electrochemical stability, and is used for electrocatalytic and photocatalytic applications [43]. Similarly, crawfish shells were used to dope porous carbon with the N atom. Doping with N increases more binding sites, which allows anchoring with Co3O4. The novel N-doped porous carbon/Co3O4 nanocomposite is used for energy storage and as anode material for Lithium-ion batteries [44]. Cotton-based denim fabric waste treated with H3PO4 is pyrolyzed to obtain P and N co-doped carbon material that serves as an efficient electrocatalyst for fuel cells. While the cellulose content serves as a carbon source, the indigo carmine dye present in denim acts as a source of the N atom [45]. Similarly, silk cotton on pyrolysis with melamine and ammonium fluoride forms an N and F doped porous carbon material which is also an excellent electrocatalyst for fuel cells [46].

This approach offers the utilization of waste materials as natural precursors to synthesize useful functionalized carbon-based nanostructures. Besides being a sustainable approach, the materials synthesized using this approach are biocompatible and less toxic comparatively.

4.1.2 Facile Approach

It involves easy methods such as one-step synthesis, one-pot synthesis, simultaneous and in-situ functionalization methods. The reduced graphene oxide (rGO) is usually produced through the reduction of graphene with highly toxic chemical reagents. p-phenylenediamine has been proven to perform simultaneous functionalization, reduction and hybridization of graphene oxide-multiwall CNT hybrid to produce the amine-functionalized rGO-CNT hybrid in an easy method. This material adsorbs methyl violet and methyl orange dye selectively from their mixture through pH modulation [47]. The Ag/rGO composites were synthesized easily by a two-step chemical reduction. Sonication of diluted GO and silver nitrate aqueous solutions followed by magnetic stirring in an oil bath at optimum conditions yields Ag/rGO composites [48].

CNT/Ag was coated with a functional polymer, namely, dopamine conjugated synthetic heparin-mimetic polymer (DA-SHP) via a one-step approach which involves anchoring of DA-SHP which reduced Ag+ ions to Ag nanoparticles followed by self-crosslinking to form o-CNT/AgNPs/SHP. The material showed antibacterial activity towards E. coli and S. aureus. The polymer coating acts as a shield for Ag nanoparticles, providing stability and biocompatibility [49]. One-step in-situ deposition of C dots on Sr(NO3)2 through sonication and calcination at optimum conditions produced SrO-C-dots [50]. CeO2 NPs/Activated Carbon nanocomposites were synthesized simply via one-step hydrothermal reaction using Ce(NO3)3 and activated carbon derived from coconut shells. It was shown that desulfurization activity of activated carbon increased with CeO2 NPs [51]. Simultaneous carbonization and sulfonation of organic matter with sulphuric acid and organosulfonic acids at higher temperatures produce SO3-H containing functionalized carbon materials in a single step [52]. Carbon dots were synthesized in-situ on amino-functionalized ordered mesoporous silica by a fast and facile microplasma-assisted method. The luminescent CDs/SBA-NH2 nanocomposite synthesized by this method showed excellent selectivity and removal of uranium ions [53]. A facile method was proposed to synthesize Ni(OH)2/CNFs composite material wherein carbon nanofiber was first treated with HNO3 followed by sonication and hydrothermal treatment with nickel precursor using an autoclave. The novel nanocomposite is used as an electrode for improving NiZn battery life [54].

4.2 Properties of Functionalized Carbon Nanostructures

The main advantage of carbon-based nanostructures is that they allow the functionalization of them with other nanomaterials or bulk materials either through covalent or non-covalent methods. The main aim of functionalization is to alter or enhance the properties of carbon nanostructures. Carbon nanostructures are mostly functionalized with nitrogen, iron, gold and silver-based or multiple functional moieties. Various properties in terms of environmental remediation, including stability, biocompatibility, electronic properties, electrochemical properties, photocatalytic and adsorption properties are modified by the material it is functionalized with.

4.2.1 Nitrogen Group Functionalized Carbon Nanostructures

Carbon nanostructures readily allow the functionalization of nitrogen-based functional groups such as amines, imines, amides and amidoxime due to their high bond strength with nitrogen. Functionalization of carbon-based nanostructures with nitrogen mainly alters the electronic properties of the pristine carbon material. N-doping alters the surface charge density due to the electronegativity difference between carbon and nitrogen atoms. This offers more catalytic active sites on carbon material and Lewis's basicity to the material.

For example, doping carbon materials with chitosan-based nitrogen imparts greater electrochemical stability and basic character. Chitosan-based N-doped carbon material has been used for electrocatalytic and photocatalytic applications. The electrochemical activity of N-doped carbon materials is influenced by the nitrogen content, surface area of the catalyst and the carbonization temperature during the synthesis of the material. Chitosan-based N-doped graphitic carbon nanomaterials showed enhanced electrocatalytic performance compared to N-doped carbon nanomaterials for the oxygen reduction reaction (ORR), which is due to the increase in specific surface area and C-N bond configurations [43]. Functionalizing the amine group on the graphene oxide-carbon nanotube hybrid offers excellent selectivity towards adsorption of cationic and anionic dye pollutants based on pH modulations. Cationic methyl violet (MV) dye is selectively removed by the rGO–CNT–PPD hybrid as a result of electrostatic interaction between the nitrogen atom of the functional group, p-phenylenediamine (PPD) of the adsorbent and the cationic dye at pH 7, while the anionic dye methyl orange

(MO) is removed at pH 3 as a result of electrostatic interaction between protonated amine functional group of PPD and the adsorbate MO [47].

Further, selective sensing of SO_2 gas has been reported for the amino-functionalized metal-organic framework by a luminescent turn-on effect. In the presence of sulphur and its derivative gases, the amino group significantly forms a complex with SO_3^{2-} ions, which hinders the LMCT effect of the amino group and the metal-organic framework, resulting in a luminescent turn-on effect [55]. Hollow carbon spheres already possess high adsorption capability because of their enlarged surface area. Further functionalization with amidoxime groups allows efficient removal of toxic uranium ions from wastewater by forming complexes through oximation and chelation reactions [56]. Doping nitrogen atoms on carbon nanotubes has been reported to show long-term stability of the material and super capacitance performance due to their excellent rate capability. However, the textural properties of CNT decreased on doping with nitrogen [57]. It has also been reported that doping nitrogen on porous carbon enhances its electrochemical performance. The highly stable C-N bond creates more active binding sites for the enhanced deposition of Co_3O_4. Nitrogen-doped porous carbon with Co_3O_4 shows good conductivity, low contact resistance and good stability, which makes it an excellent anode material for lithium-ion batteries [44]. Nitrogen functionalized ordered mesoporous carbon has been used as catalyst support to disperse and stabilize small-sized Pd nanoparticles due to their enhanced physicochemical performance. The N-doping also enhances catalytic activity and selectivity for catalytic hydrogenation of phenol [58]. Similarly, the Pd catalyst supported on N functionalized activated carbon showed enhanced catalytic activity for nitrite and bromate hydrogenation [59].

Thus, functionalization of carbon nanostructures with nitrogen enhances stability, selectivity, electronic, catalytic, electrochemical, and physicochemical properties.

4.2.2 Iron-Based Functionalized Carbon Nanostructures

2D-Carbon flakes allow facile functionalization of the surface with Fe_3O_4 nanoparticles. Fe_3O_4 is easily available at a low cost and imparts excellent magnetic properties and biocompatibility. The synergistic effect of Fe_3O_4 nanoparticles and 2D carbon flakes offers stability to the material and adsorption properties are increased [35]. Similarly, the stability and adsorption properties of the carbon sphere are enhanced by encapsulating iron nanoparticles (nZVI) due to the synergetic effect on functionalization [60]. Functionalization of the multi-walled carbon nanotube with the iron oxide coating on its surface enhances the sorbent properties, useful for water treatment [61]. Fe_3O_4 nanoparticles anchored on graphitic carbon nitride offered the magnetic property to the material, which then showed enhanced adsorption capacity of metal ions due to the magnetic separation process [62]. Coating iron nanoparticles with carbon also prevents the 'ageing effect.' Carbon encapsulated iron nanoparticles are quite stable for long years and prevent them from releasing the pollutant ions back to water resources [37]. Encapsulating Fe_3C nanoparticles in nitrogen-doped carbon material offered high stability to the material and enhanced electronic conductivity and enzyme-like catalytic properties [63].

Table 1 *Functional carbon-based nanostructures for environmental remediation*

S. No.	Functionalized carbon-based nanostructure	Functionalizing agent	Synthesis method	Application	Ref
1	Fe_3O_4@2D-CF composite	Ferric chloride hexahydrate	Pyrolysis	Removal of Arsenite from water	35
2	Ag/rGO	Silver nitrate	Chemical reduction	CNT IDEs-Ag/rGO as superior NO_2 gas sensor	48
3	Ag-CNT	Silver nitrate	Nano-interfacial coating	Antibacterial	49
4	Amidoxime functionalized hollow carbon sphere	Hydroxylamine hydrochloride	Oxidation	Removal of Uranium from wastewater	56
5	rGO-CNT-PPD	p-phenylenediamine	Simultaneous functionalization and reduction	Selective removal of methyl violet and methyl orange by adsorption	47
6	MOF-5-NH_2	Amino benzene dicarboxylate	Mechanical stirring	Luminescent sensor to detect SO_2 gas	55
7	CDs/Au NPs composite	Citrate stabilized Au NPs	Mechanical stirring	Colorimetric nano sensor for sensitive and selective detection of Ag+ ions in water	64

8	SrO-C-dot composite	Strontium nitrate and PEG400	Sonication and subsequent calcination at the inert atmosphere	Conversion of microalgae to fatty acids	50
9	CN-Ni$_2$P	Nickel chloride hexahydrate and sodium hypophosphite	Heating at the inert atmosphere	Transformation of microplastics into H$_2$	65
10	Carbon encapsulated iron nanoparticles (CE-nFE)	Iron(iii)nitrate nonahydrate	Hydrothermal carbonization	Heavy metal removal of contaminated water	37
11	CeO$_2$ NPs/AC	Cerium nitrate	Hydrothermal	Enhanced SO$_2$ removal capacity	51
12	CA-CDs-FF (Fluorescent film)	Chlorogenic acid (CA), Poly-vinyl alcohol (PVA)	Ultrasonication	Detection of contaminants in food	66
13	Carbon nanotube coated carbon fiber (CNTCF)	Acetylene (C$_2$H$_2$)	Chemical vapour deposition	Sensing/ detection of methanol gas	67
14	Multi-walled nanotube bucky paper	Toluene/ferrocene liquid mixture	Chemical vapour deposition	Nano-filter for gold nanoparticles	68
15	Ethyl acetate- SW-CNTs	Carboxylate functional groups	Covalent chemical functionalization	Solubilization of CNT in aqueous water and Uranyl adsorption	69
16	Oxygen group functionalized CNT hydrogel	Nitric acid and sulfuric acid	Acid treatment	The enhanced current generation in microbial fuel cell	70
17	−CO−, −C=O, and −COO− functionalized MWCNT Ag interconnects	Oxygen	Oxygen plasma treatment	Improved adhesion of Ag-MWCNT-Ag interconnects	71
18	Co$_9$S$_8$ nanorod arrays on C fiber	Cobalt hydroxide coated on eggshell membrane	In-situ carbonization and sulfurization	As electrodes in Li-ion and Na-ion batteries	36
19	Benzidine functionalized cross-linked CNT	Bisdiazonium compounds	Chemical cross-linking	Removal of organic pollutants and Separation of water/oil emulsions	72
20	CTAB functionalized ZnO/CNT on Ag films	CTAB	Sol-gel method	Quantification of catechol in wastewater	73
21	Cu/CNT membranes	Cu NPs	Magnetron sputtering	Removal of Arsenite through microfiltration	74
22	EDTA functionalized Activated Carbon	TMS-EDTA	Chemical treatment	Adsorption of rare earth from aqueous solution	75
23	Fe$_3$C/N- C nanofiber	Polyacrylonitrile and Iron (III) nitrate mixture	Electrospinning	Colorimetric assay for determination of phenol	63
24	Graphitic carbon nanoparticles	Sesame oil	Pyrolysis	Removal of pollutant dyes from water by adsorption	76
25	MgAl-NO$_3$-LDH	Citric acid and urea	Co-precipitation	Adsorption of SeO$_4^{2-}$ and Sr^{2+}	77

26	Iron oxide coated MWCNTs	$FeCl_3$, $FeSO_4$	Microwave-assisted process	Removal of trace level Arsenic from water	61
27	Fe_3O_4-g-C_3N_4	An aqueous solution of FeCl3: FeSO4	Ultrasonic method	Removal of zinc, lead and cadmium ions from water	62
28	NH_2 functionalized mesoporous silica carbon dots	Citric acid and ethylene diamine	Microplasma-assisted method	Selective removal of Uranium ions from water	78

5. Conclusion

This review represents an overview of various carbon nanostructures and how their properties are enhanced upon functionalization for environmental remediation. We have discussed the sustainable and facile functionalization aspects, literature review on nitrogen and iron functionalized carbon nanostructures, roles of functionalizing moieties towards potential water treatment, toxic gas detection and electrode materials. The importance of sustainable synthesis of functionalized carbon nanomaterials from green waste materials and their stability properties are highlighted. Functionalized carbon materials synthesized by eco-friendly methods offer high stability and less toxicity and are highly preferred for applications in environmental remediation. Hybrid composite structures of carbon nanoallotropes show great opportunities for exciting properties and wider applications in future. Therefore, the growth of research in this area is expanding. However, sustainable synthesis of carbon nanostructures from biowaste beyond laboratory scale is a huge challenge and therefore commercialization of which is necessary for real-time applications.

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Conflict of Interest

Authors declare that there is no conflict of interests regarding the publication of the paper.

Author Contribution

The authors confirm contribution to the paper as follows: ***study conception and design:*** *Badal Kumar Mandal;* ***data collection:*** *Badal Kumar Mandal & Nishanthi Vasanthi Sridharan;* ***analysis and interpretation of results:*** *Nishanthi Vasanthi Sridharan;* ***draft manuscript preparation:*** *Nishanthi Vasanthi Sridharan. All authors reviewed the results and approved the final version of the manuscript.*

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