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Review

Processing of Carbon-Based Nanomaterials for the Removal of Pollutants from Water/Wastewater Application

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Abstract: In both the inorganic and organic worlds, carbon-based nanomaterials, such as benzene, diamond, graphite, fullerene, and carbon nanotubes, are abundant. In science laboratories, carbon is the focal point of activity. In this overview, the synthesis, characteristics, and several uses of graphene—including energy conversion, energy storage, electronics, and biosensing—were explored with a focus on ecologically friendly production techniques. This article also discusses recent advancements in the detection and treatment of organic contaminants and heavy metals utilizing nanomaterials. In this article, we outline some recent developments in the creation of innovative nanomaterials and nanostructures and methods for treating organic contaminants and heavy metals in water. The essay presents the current state of the field and, in our opinion, should be helpful to anybody interested in nanomaterials and related materials.

Keywords: carbon-based nanomaterials; removal of pollutants; heavy metals; wastewater application

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Citation: Singh, R.; Samuel, M.S.; Ravikumar, M.; Ethiraj, S.; Kirankumar, V.S.; Kumar, M.; Arulvel, R.; Suresh, S. Processing of Carbon-Based Nanomaterials for the Removal of Pollutants from Water/Wastewater Application. *Water* **2023**, *15*, 3003. https://doi.org/10.3390/w15163003

Academic Editor: Naresh Singhal

Received: 21 July 2023
Revised: 8 August 2023
Accepted: 13 August 2023
Published: 20 August 2023

1. Introduction

Two-dimensional graphene has a hexagonal arrangement of carbon atoms, where carbons hybridize to form three powerful C-C bonds that are 120° apart. The identical p-orbitals are conjugated on additional carbon atoms so that they are parallel to the sp^2 hybridization plane [1,2]. The electrical and mechanical characteristics of graphene are distinctive. Graphene has an unusual zero-gap semiconductor capabilities with a charge carrier mobility of $>2 \times 10^5$ cm^2/Vs at an electron density of 2×10^{11} cm^{-2} and a thermal conductivity of >3000 W/mK because its valence band mirrors its conduction band, with the two bands crossing at the Fermi level. Graphene has a potential surface area of >2500 m^2/g due to the carbon atoms being completely exposed in a planar manner [1,2]. In order to create composites, graphene has been combined with metals, alloys, oxides, and other polymeric materials [1,2]. The addition of heteroatoms to the graphitic plane can further alter the characteristics of graphene. The electrical neutrality of the graphene network is compromised when a carbon atom is swapped out for another one, which increases the susceptibility of the 2D network to chemical reactions. In-depth research has been carried out on novel features of graphene and its hybrid structures for cutting-edge technological applications in electronics, optics, catalysis, and energy storage. Graphene, which has just emerged as a novel class of carbon nanostructures, has garnered a lot of attention and is now a fast-growing field. It exhibits a wide range of features, including thermal conductivity, charge carrier mobility, electrical and mechanical stability, magnetism, and more. These characteristics, along with a high surface area, are essential in electrochemical, optoelectronic, and medicinal applications. Recent attention on graphene as a generic foundation for diverse composites has, most crucially, sparked a lot of potential in the fields of energy and the environment [3,4]. However, due to (i) graphene's poor light absorption and low capacitance, (ii) its ease of stacking and agglomerating in solvent, and (iii) its nature as a zero-gap semi-metal, two-dimensional graphene sheets are restricted in many specialized domains. Growing efforts have been made to modify the graphene surface and create dimension-tailored functional graphene structures, such as graphene quantum dots (QD), graphene fibers (1D), graphene sheets/films (2D), and graphene gel (3D), in order to increase the range of applications in areas such as quantum computing, catalysis, sensors, and more [5–8].

Among these, macroscopic materials based on graphene with a three-dimensional (3D) porous network have drawn more interest in the fields of energy and the environment. Graphene-based 3D materials provide greater benefits than carbon-nanotube-based 3D designs, including simple preparation, high efficiency, and affordability. 3D graphene-based materials may be created by combining separate 2D graphene sheets that have undergone chemical modification into monoliths that have 3D microporous features [8–11]. This study aims to provide an overview of current advancements in graphene, doped graphene, and its composites with nanoparticles (NPs) used in chemical, material, and energy applications. Figure 1 demonstrates the conventional technologies used for wastewater treatment and their shortcomings. Nevertheless, the adsorption method is advantageous because it is flexible, economical, operational, and simple and no secondary contamination is generated during the process. To emphasize the crucial elements of graphene chemistry and its different uses, this article contains both the work of the authors' groups and typical examples from other fields. An appropriate setting is provided to explore the synergistic impact of graphene and NPs on increased catalytic capabilities due to the more controlled production, which results in high-quality graphene and its solutions in diverse solvents. The review will concentrate on the uses of nanomaterials based on graphene in electrochemical energy storage and conversion systems.

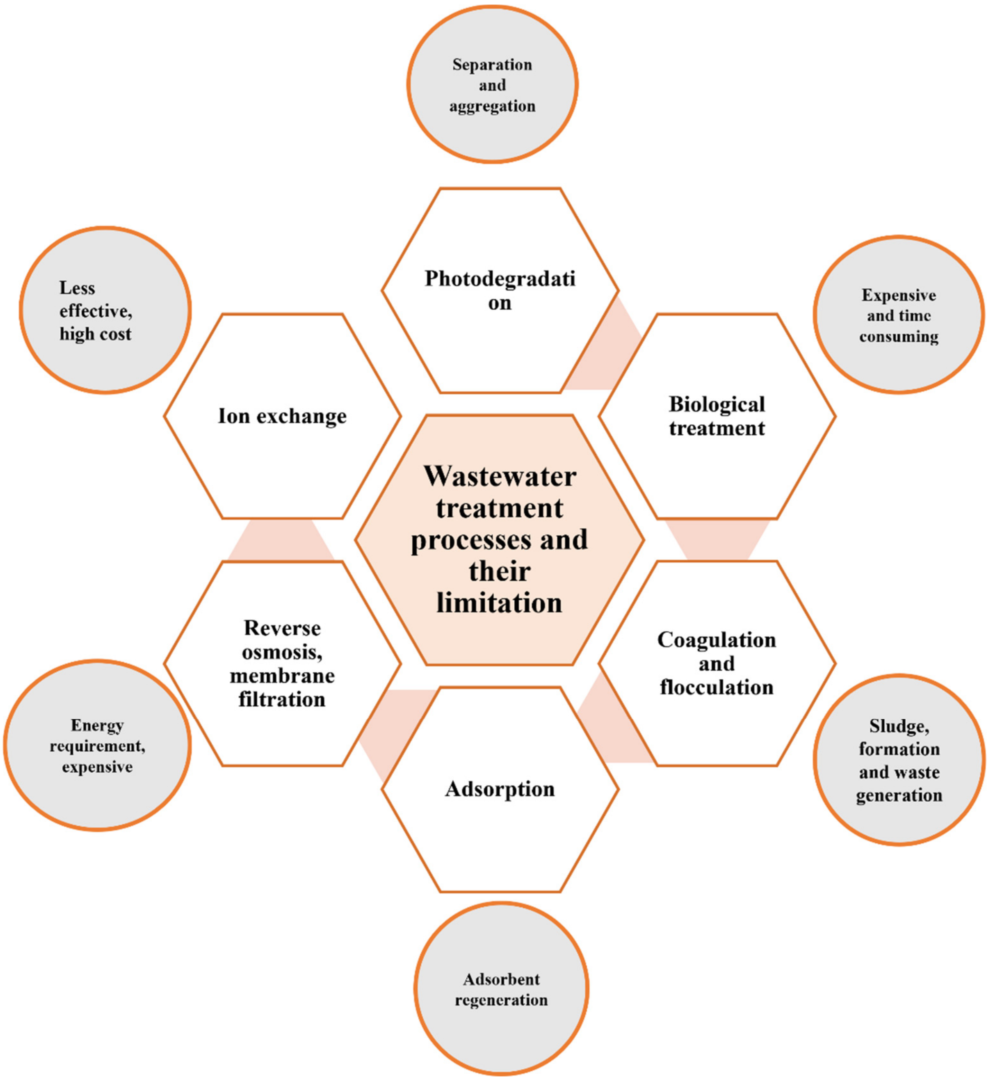

Figure 1. Scheme illustrating conventional wastewater treatment and its drawbacks.

2. Application of Graphene Materials in Antibiotic Removal

Graphene adsorbents are essential for the filtration of water. There are several forms of graphene allotropes (Figure 2). Multiple studies have shown that pollutant adsorption onto the surface of the photocatalysts is enhanced by graphite materials. Additionally, heteroatoms such as B, N, and S are capable of being employed to change electronic characteristics to increase photocatalytic operations, accelerate electron transport, and offer additional active sites. The presence of carboxylic, amino, hydroxyl, and thiol groups that function on the external layer of N/S-doping-based carbon aerogel aids in the elimination of organic contaminants from water. As a result of its surface polarization, graphene can react to substances such as antibiotics [1]. Graphene has just a single film of C-atoms, meaning that the atoms are entirely exposed to its surroundings and might potentially have an interaction with substances such as antibiotics. Given its larger surface area as well as porosity structure compared to normal adsorbents, graphene has recently been offered as a viable candidate for speedy binding to antibiotics, resulting in fast adsorption [2]. Many antibiotics may be degraded using graphene, including graphene composites. Zhang et al. [12] examined the effects of graphene with an Fe_3O_4 composite on the degradation of tetracycline, an antibiotic, in aqueous solutions. It seems the mixture may be a deteriorating substance. Positive findings were seen for antibiotic destruction when a composite made up of $MnFe_2O_4$+C_3N_4+ graphene was used in another study. When testing the elimination of metronidazole from a combination of four (tetracycline, amoxicillin, and ciprofloxacin) medications, the $MnFe_2O_4$+C_3N_4+ graphene composite was around 3.5 times more effective than the original g-C_3N_4. Table 1 shows different types of graphene-based nanomaterials, antibiotics, and mechanisms of adsorption.

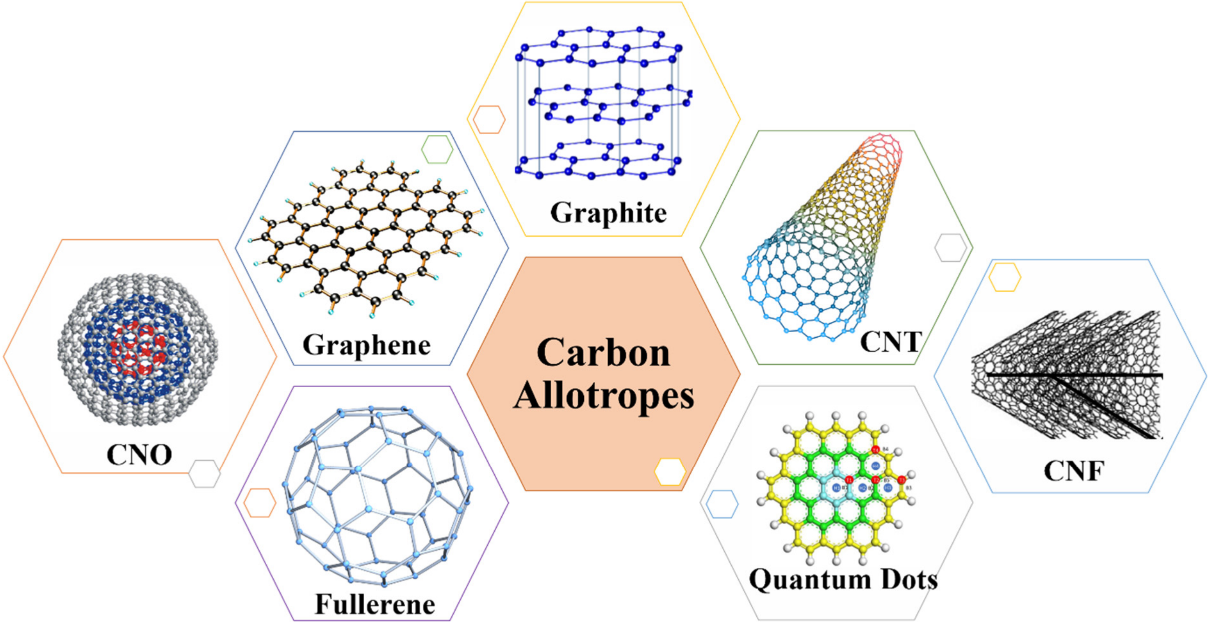

Figure 2. Various forms of carbon allotropes and their derivatives.

Table 1. Different types of graphene-based nanomaterials, antibiotics, and mechanisms of adsorption.

S.No	Type of Graphene-Based Nanomaterial	Antibiotic/Heavy Metal	Mechanism	References
1	Magnetic graphene oxide sponge	tetracycline	Adsorption	[4]
2	Fe_3O_4@TiO_2-GO	Enrofloxacin	Adsorption	[5]
3	GO@Fe_3O_4/ZnO/SnO_2	Azithromycin	Photocatalytic degradation	[6]
4	rGO@*n*Fe/Pd	Rifampicin	Fenton-oxidation	[6]
5	CdS–Bi_2MoO_6/RGO	Ciprofloxacin	Photocatalytic degradation	[7]
6	Graphene-phase biochar	Tetracycline	Adsorption	[10]
7	Graphene–chitosan (GO-CS)	Chromium (VI)	Adsorption	[13]
8	GO-CS@MOF [Zn(BDC)(DMF)]	Chromium (VI)	Adsorption	[14]
9	β-cyclodextrin-modified graphene oxide (β-CD-GO)	Cadmium	Adsorption	[15]
10	chitosan/graphene oxide composite	Chromium (VI)	Adsorption	[16]

As a variant of graphene oxide (GO), reduced graphene oxide (rGO) is classified as a useful polymer-based filling composite substance because of its superior properties and ease of use. Pristine graphene is demonstrated to be moderately dispersive in water-based solutions because of its tiny gaps in between the layers and hydrophobic characteristic [2]. Its effectiveness and usefulness increase as it is reduced or oxidized. Ersan et al. agree that graphene is the most promising of the next generation of adsorbents because of its high adsorption selectivity for organic substances with open-up sheet form. Organic contaminants and GO have been demonstrated to interact strongly with each other. Tetracycline has

-NH_2 groups, which have an exceptionally high electron density, making it a potent electron giver. Several rGO composites have been shown to have antibiotic-remediating properties. Antibiotic ineffectiveness is affected by several variables. The π-π interaction between rGO and antibiotics has been hypothesized to improve antibiotic adsorption. Fewer antibiotics may be absorbed by a material with a larger surface area as a greater number of sites for adsorption is available. Shen et al. found that a composite weighing 0.005 gm made up of Cu_2O, rGO, and Bi_2O_3 degraded tetracycline at 80%. Antibiotic degradation uses nearly double the quantity of rGO + Cu_2O and quadruple the quantity of rGO + Bi_2O_3 [17].

The composite MoS_2/Cu_2O/rGO was synthesized by Selvamani et al. [18] for the aim of tetracycline breakdown. Following 2 h of interaction, the TiO_2-rGO nanocomposites successfully decomposed ibuprofen, sulfamethoxazole, and carbamazepine. When compared to raw TiO_2, TiO_2-rGO showed significantly increased degradation by photocatalytic reactions. The degradation rate was very sensitive to the rGO content of the catalysts. Biochar-based graphene substances [8] have shown promise for removing antibiotics from water. Antibiotic adsorption was improved in biochar-based composites due to the presence of GNMs. Their techniques have been shown to be efficient by multiple studies. Huang et al. investigated the effect of biochar derived from bamboo dust with GO on the breakdown of sulfamethazine [9]. Sulfamethazine adsorption has been shown to be two times greater with activated biochar compared to pure biochar. It is expected that both the surface area and the amount of oxygen functional groups located on the surface of the substance increase after incorporating GO. In an investigation evaluating different pH levels and ionic strengths, it was discovered that antibiotic adsorption was enhanced by lowering the ionic strength. The optimal range for adsorption was identified as 3–6 on the pH scale. After four regeneration cycles, the adsorption capability of rGO + biochar produced from maize stems for atrazine adsorption was 55 mg/g. Improved oxidation systems will continue to evolve if they are both reusable and reliable [11]. Keeping an impermeable barrier between the produced material and the liquid solution is essential at this point [11].

Separation using magnetic substances saves time and money compared to the conventional technique [19]. Magnetic composites can be utilized to get around the inherent limitations of photocatalysts and adsorbents by facilitating quick separation after adsorption as well as the photocatalytic process and allowing further recycling. Magnetite (Fe_3O_4) has been demonstrated to be the most likely nanoparticle that has been researched so far. These nanoparticles are ideally suited for destroying or adsorbing their target molecules due to their high surface area and strong magnetic properties [19]. Either as a catalyst or an adsorbent, the synthetic magnetic composite is used to remove organic and inorganic contaminants from water and wastewater [20]. The production of active photogenerated entities, such as ozone, hydroxyl, and electrons, has resulted in excellent catalytic properties and remarkable versatility when used for pesticide treatment in water-based solutions, as is the case with magnetic composites [20]. The produced photocatalyst was used in a study by Al-Kahtani [20] for up to seven cycles with constant catalytic activity. Thus, it has been demonstrated that the photocatalyst/adsorbent value increases with the number of times it may be reused.

3. Graphene-Based Adsorbents for the Sorption of Organic and Heavy Metal Pollutants

Medications and aromatic compounds, pesticides and herbicides, and antibiotics are just some of the organic pollutants that may be removed from water using adsorbents based on graphene (Figure 3).

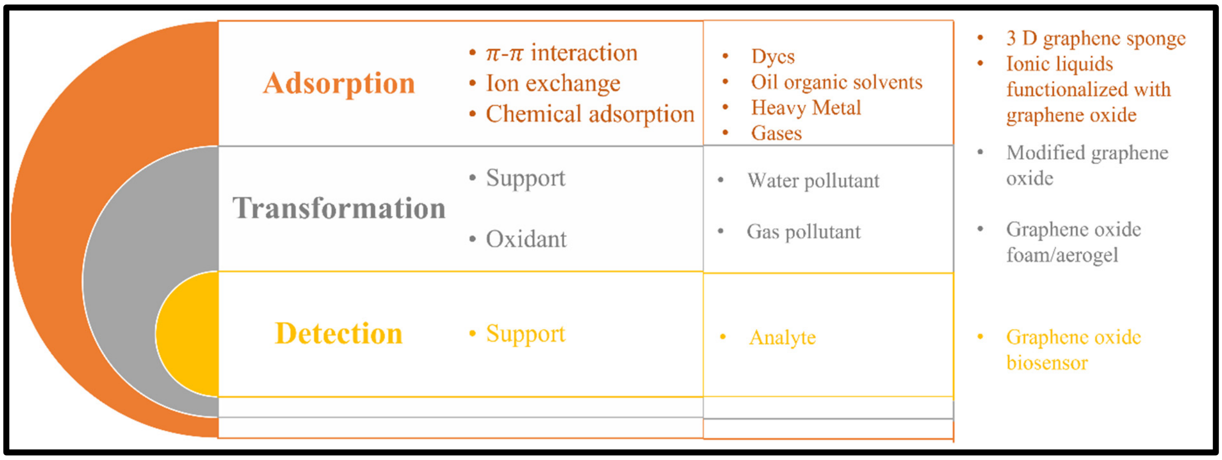

Figure 3. Scheme illustrating graphene properties and its application in organic and heavy metal pollutants.

Industrial activity, such as drug manufacturing, agrochemical processing, mining for coal, and farming, all add to the contamination crisis. Such pollutants are harmful to the endocrine system as well as the physicochemical properties of water [21], which can also cause cancer. Here, further research is continuing to be carried out with the aim of eliminating additional organic pollutants with graphene composites, as has already been reported by various researchers. Water contaminants such as carbamazepine (CBZ), bisphenol A (BPA), acetaminophen (ACP), caffeine (CAFF), and triclosan (TCS) were eliminated with the use of a composite made of activated carbon, GO, and chitosan [19]. The Langmuir model provided a perfect match to the isotherms, which showed the highest absorption values for such pollutants. Researchers discovered that a polar solvent resulted in ~70–80% desorption of BPA, CAFF, and ACP, whereas a nonpolar solvent resulted in ~60% desorption of CBZ and TCS. Recently, researchers developed a noncovalent GO-based composite functionalized using the ionic liquid 1-hexyl 3-decahexyl imidazolium for the uptake of carbamazepine (CBZ), sulfamethoxazole (SMZ), and ketoprofen (KET) [21–23]. The absorption capabilities were 58, 89, and 84 mg/g after 10 min of absorption. The sorption results were satisfactorily explained by this Langmuir model, suggesting that single-layer adsorption took place. The researchers reported that the removal percentage was 63% for KET, 75% for SMZ, and 58% for CBZ with seven cycles of desorption utilizing ethanol/methanol as the dissolving solvent. Using a ternary compound composed of rGO, maltodextrin, and CuO_2, Moradi et al. were able to eliminate pharmaceuticals including amoxicillin and diclofenac, with absorption efficiencies of 12.9 and 12.8 mg/g, respectively [24]. The authors concluded that chemisorption along with monolayer adsorption of molecules occurred as their results were consistent with the PSO as well as Langmuir models. After reusing the material as an adsorbent with 1 M NaOH as the eluting agent, the research team found that the adsorption capability for TC dropped from 306 to 227 mg/g [24]. A novel magnetic nanoadsorbent consisting of Fe and Co nanoparticles paired with GO was employed by investigators at the University of Brasilia to remove the pharmaceutical contaminant caffeine in water. The maximum caffeine absorption was 29 mg/g at a starting dosage of 10 mg/L [24]. The natural procedure generated heat and had a positive thermodynamic outcome [24]. The researchers claimed that the main adsorption technique was the formation of hydrogen bonds connecting the nitrogen atoms that make up the amino groups of the caffeine and the hydrogen atoms of the hydroxyl groups containing -OH of the GO, with GO, Fe, and Co also interacting with electrons that reside on the caffeine [24]. In order to efficiently remove doxycycline from waterways, Xiong et al. created a charcoal-based GO composite that is reusable, robust, and adaptable [25]. Its maximum absorption was 797 mg/g, and the sorption data fit the Langmuir isotherm model. The researchers claimed that hydrogen bonding, electrostatic attraction, and stacking were primary mechanisms in doxycycline adsorption on the BC-GO composite. Januário et al. synthesized adsorbents to facilitate absorbent elimination in water containing COVID-19 treatment drugs such as dipyrone and chloroquine [26]. The highest amounts of such drugs that could be taken up by the BC-GO adsorbent were 62 and 37 mg/g. This research confirmed that adsorption is a process that is endothermic according to the Langmuir model. In addition to testing, it as an adsorbent on laboratory wastewater, researchers were able to remove pollutants from wastewater with an 82% effectiveness rate. The authors hypothesized that adsorption is controlled by π-π interactions and H-bonding because of ionic as well as pH variations [26]. De et al. [27] produced a unique membrane for absorbing propanol drugs with an accumulation capability of 200 mg/g, which consisted of hydrolysis polyacrylo-nitrile, including GO enhanced with safranin. Multilayered adsorption on a heterogeneous surface was revealed by the data's fit to the Freundlich model [27]. Adsorption was discovered to include cation substitution, electrostatic connections, and π-π interactions. Liu and team utilized a chitosan composite of GO magnetic adsorbent to remove sulfo-salicylic acid in water [28]. This process was exothermic in nature and governed by entropy. The adsorbent was utilized three times in 10 mmol/L. The solution of sodium hydroxide resulted in a rate of removal of 96.89%. Many potential mechanisms were proposed, including differences in skin color, hydrogen bonding, beta–gamma interactions, and even electrostatic forces. By integrating beta-cyclodextrin-confined rGO by MW-CNTs, Feng et al. [29] developed a novel adsorbent that allows effective elimination of naproxen in water. The outcomes were compatible with multiple layers of adsorption and the Freundlich isotherm model, with the highest absorption at 132 mg/g. The research team hypothesized that naproxen adsorption within the pores of beta-cyclodextrin was due to the hydrophobic interface among the adsorbent as well as electrostatic attraction, H-bonding, and π-π interactions. Perfluorooctanoic acid (PFOA) was removed from water using GO aerogels [30]. The aerogels had a PFOA removal efficiency of 1574.8 mg/g. The Freundlich model, which accurately describes the phenomenon, anticipated the presence of a heterogeneous adsorbent layer [30].

Industrial polluting sources, such as plating, painting, mining, metal smelting, automobiles, refineries, and many more, contaminate numerous bodies of water with heavy metal ions, including Cr, Fe, Pb, Ni, Hg, Cd, As, etc. Overexposure to these harmful metal ions impact humans as well as aquatic creatures. Graphene is often used to remediate heavy-metal-contaminated wastewater [30]. Madadrang and others [31] improved GO by EDTA–silane for Pb (II) absorption. The highest ability was 524.5 mg/g based on PSO kinematics as well as the Langmuir isotherm model. Another researcher, Verma [32], coprecipitated magnetic hybrids of GO (GO@$MnFe_2O_4$) to increase absorption of As (V), Pb (II), and As (III) from water. The removal process was highly dependent on the outside temperature and concentration, and results showed the absorption was best at pH 5. The PSO and Langmuir models of adsorption agreed with maximum absorption rates of 207, 672.8, and 146.2 mg/g for As (V), Pb (II), and As (III), respectively [32]. A novel adsorbent was created through the integration of Fe_3O_4 nanoparticles on top of a GO–chitosan composite to remove ions of Cu (II) and Cr (VI) from water-based solutions. Their absorption capacities were 143 and 111 mg/g, respectively. The solid-solution interface was endothermic and stochastic. Cr (VI) sorption was strongest at pH 3 because larger pH values enable more hydroxyl (OH) ions in the solution itself and compete with the positively charged interface for sorption [32]. Kong and the research team created a triad GO-NH_2-HBP-CMC nanocomposite to enable removal of heavy metals from water [33]. The Langmuir model matched the real adsorption, suggesting monolayer sorption. Pb^{2+} and Cu^{2+} showed the greatest sorption capabilities with 153 and 137 mg/g, respectively. The authors suggested that external and pore diffusion had influenced sorption. The main chemical reactions were metal ion exchange involving O groups and complexation with N groups. By encapsulating silica on GO along with functionalizing using chitosan, the newly designed composite GO-Si-CS adsorbed Pb (II) from a water-based solution [33]. The study further claimed that both multilayer adsorption and chemisorption occurred during the procedure. The

Langmuir model showed 256 mg/g as the maximum capacity for absorption. GO-Si-CS with interaction surface functional groups were essential for Pb (II) removal [33]. Recent research showed that polypyrrole-based cobalt oxide GO could eliminate Pb(II) from an aqueous solution. Batchwise sorption tests showed maximum Pb (II) and Cd (II) absorption of 780 and 794 mg/g, respectively [34]. The optimal batch adsorption parameters were pH of 5.4 for Pb (II) and pH of 6.0 for Cd (II) at 50 °C, 0.33 g/L of adsorbent, and 250 or 150 min. of contact time. The Langmuir isotherm as well as the PSO model suggested single-layer adsorption along with chemisorption. Thermodynamic studies proved the processes were spontaneous and endothermic. With acidic pH, the composite with GO + OH + COOH were protonated and positively charged, pushing out positive Pb^{2+} ions and reducing adsorption. Deprotonation increased pollutant adsorption on the surfaces. However, above pH of 5.4 and 6.0, the abovementioned ions precipitated as $Pb(OH)_2$ and Cd (OH), thereby lowering the efficiency. Attia et al. developed an antimicrobial graphene + polypyrrole nanocomposite for pH-dependent aqueous Mn^{2+} absorption [35]. The Langmuir model was well satisfied, revealing that monolayer adsorption occurred at 56 mg/g at pH of 5.5. Ranjan Rout [36] developed a graphene composite to remove Cr (VI) from water using an adsorbent weighing 5 mg in a 50 mL solution (i.e., 100 mg/L) with Cr (VI) concentration of 1321 mg/g and pH 1. The adsorption results fit the Langmuir isotherm and the PSO kinetic model. Perlova et al. (2020) [37] studied a composite based on zirconium hydrophosphate, which allowed practically complete removal of U(VI) from water in the presence of hardness ions; the highest regeneration degree was also achieved.

4. Applications of Graphene Based Membranes in Water Purification

Covalent crosslinking through esterification methods increases the permeability of GO membranes. By employing aqueous solutions containing equal parts KCl, $CaCl_2$, $MgCl_2$, and $NiCl_2$, the linked GO membranes exhibit higher fluxes during permeation compared to the pure GO membrane. Jia and Wang [38] measured a K^+/Mg^{2+} sensitivity of 6 using this membrane. CG membranes (crosslinked graphene) as well as traditional UF membranes made of polymers were used to segregate organics, with a purity of 99.9% and a flow rate of 225 LMH ($L/h/m^2$). The RGO membrane improved the water permeability to 61.7 LMH under partial reduction environments. Nevertheless, by creating an RGO membrane that had higher than normal reduction circumstances, an elevated salt rejection percentage was achieved (81% of Na_2SO_4). A new membrane of GO + crosslinked polyimide was created using simple pressure-assisted filtration for utilization in organic solvents [39]. The resulting GO/CLPI membrane increased the filtering ability for Rose Bengal salt molecules to >94%, with an increase in solvent flow of 11, 4.9, and 1 LMH for water, IPA, and DMF, respectively. Adding warmth enabled the GO film to contract completely, producing a membrane with a high fouling resistance (FRR) of >63. Following that, Li et al. [40] created a solvent-resistant (SRNF) membrane based on thin-film nanocomposites (TFNs) containing amino-functionalized graphene-based quantum dots (a-GQDs). The generated membrane had an increased ethanol permeation of 32 LMH, or 44% above the pure membrane, due to the flaking morphology of the a-GQDs, which can provide greater openings for solvent absorption. In contrast, the percentage of cells that opposed rhodamine-B (RDB) was not altered over time. The resulting Rose Bengal membrane was 99% pure after 768 h, demonstrating remarkable resistance to the solvent. The antifouling performance was improved by developing a borate inorganic crosslinked long-lasting GO membrane [40]. Extensive washing tests with NaOH + sodium dodecyl sulfate revealed that the resulting inorganic crosslinked GO membrane maintained its rejection percentage, flow, and overall structure. Organic fouling was successfully decreased by the inclusion of trace -OH radicals. Therefore, a novel, long-lasting crosslinked GO membrane was demonstrated for fouling prevention during the treatment of wastewater. The antimicrobial membrane of dopamine with polyethyleneimine crosslinked GO showed over 99% antibacterial efficiency while illuminated with a 795 nm NIR laser [41]. The photothermal response and the antibacterial potency of the GO-PDA-PEI membrane were also unchanged during five cycles of reuse and recycling. At just 1 bar, the PPD NF membrane was able to extract 99.9% of methylene blue (MB). As pointed out by Pandey et al., the PPD crosslink additionally enhanced the longevity, reuse ability, and sturdiness of the GO membrane [42,43]. In spite of the porosity of the hydrogel created by the crosslink, the three-dimensional hydrogel GO membranes demonstrated an average water flow of 111.5 LMH, which was 14 times larger compared to that of an untreated GO membrane. The rejection rate for MB in GOMHs was 98%, which was greater than that of the pure GO membrane due to the enhanced interconnection of the tiny pores combined with the ability to adsorb the hydrogel structure. Further NaCl was rejected by the PEI crosslinked GO membrane compared to the pristine GO membrane. The EDA-GO membrane was shown to have a significant rate of retention of >95% in dye soaking in methanol. It could let 14 LMH of water through it (pure GO allowed 9 LMH). An MXDA crosslinked GO sheet partially interpenetrating the PSf substrate exhibited comparable dye rejection of >99% as well as a permeance of 4 LMH [43]. The copper crosslinked GO membrane's two-dimensional nanochannels were clearly defined and stable, allowing effective molecular segregation. Using a K+ infiltration efficiency of 0.174 mol m^{-2} h^{-1} and a K^+/Mg^{2+} extraction factor of 68, this membrane demonstrated outstanding results for mono/multivalent metal ions sifting. The amino acids in crosslinked membranes had a permeability of 12 LMH at room temperature and pressure. The removal coefficient of this membrane was 94% for alkali metals and 96% for alkaline earth metals. Furthermore, the composite membranes' excellent antibacterial properties against E. coli could be attributed to the razor-sharp edges of the sheets of graphene, which encouraged membrane strain for structural deterioration of the cell membrane. Na_2SO_4 rejection percentages of 58%, 74%, and 82% were achieved by the pure GO, GO-EDA-V, and GO-EDA membranes, respectively. The water absorption rates were 11, 2.2, and 2.3 LMH. Because EDA's covalent crosslinking prevents the hydrating of nanosheets made of GO and restricts the growth of interfacial space, EDA-GO membranes allow less penetration and more rejection. EDA membranes have been proven to exhibit rejection rates of more than 99% in optimum separating conditions. Furthermore, the rejection rate for Cr (VI) by GO/EDA membranes was greater than 99% in acidic environments due to the reduced d-space and increasing ion size [43]. Compared to organic nanofiltration, the present GO-based membranes, and two-dimensional MoS_2-based membranes, the salt rejection (NaCl) of GOF membranes crosslinked with TU was 96% [41]. Regarding the process of treatment of the modeled oilfield wastewater, TA + GO membranes lowered chemical oxygen demand by 91.7% and rejected total dissolved solids and turbulence with high efficacy while not affecting their mechanical strength. The PDA-GO membrane had better flow performance despite having a biofilm layer that was 45% lower compared to that of the pure membrane [44,45]. After 48 h of operation, the PDA-GO membrane lowered the flux by 33%, whereas the unmodified membrane lowered the flux by 55%. PDA-GO's powerful antibacterial activities can be attributed to the membrane's aqueous and smooth outer layer. The separating factor of GO-PVAm-silica membranes for butanol dehydration reached 1189, which was approximately 2.5 times greater compared to that of GO-PVAm membranes, whereas the flow rate was ~13 kg m^{-2} h^{-1} [41]. According to research, the modified membrane was able to eliminate NaCl at a rate above 99.99%. The Zm-PEI-GO@PDA/PESn NF membrane demonstrated a flow rate of 49.55 LMH and a deletion amount of 100% for Congo Red, 83% for Orange G, and 66% for methyl orange [46]. By forming a dense hydration barrier and preventing protein adhesion, the patched zwitterionic polymer enhanced the membrane's ability to resist fouling resistance [39]. Na_2SO_4 removal efficiency was 71.8% at an average flux of 33 LMH through a PDA-GO/-CD-EDA membrane. Drugs such as carbamazepine were rejected more effectively at lower pH and tighter d-space. Oil–water mixtures with a maximum flow rate of 119,425 LMH were found to have a removal effectiveness higher than 99% [47]. Recent research by experts showed that carbon aerogels are a fascinating three-dimensional (3D) monolithic porous material that can be widely applied in environmental chemistry for removing pollutants such as oils, toxic organic solvents, dyes, and heavy metal ions in aquatic environments [48].

Researchers have shown that rGO/CuO nanocomposites can be used for dye degradation of organic pollutants in wastewater remediation [49]. Efficient removal of heavy metals and organic pollutants from aqueous solutions can be carried out using graphene oxide nanocomposite [11,13–15,50–54].

5. Application of Graphene-Based Materials in EMs

Fireworks, explosives, propeller fluid all make use of energetic materials (EM) [55,56], which are a category of mixtures that contain chemicals that are flammable with a significant chemical interaction and generation of energy. There are several forms of graphene oxide and its derivatives (Figure 4).

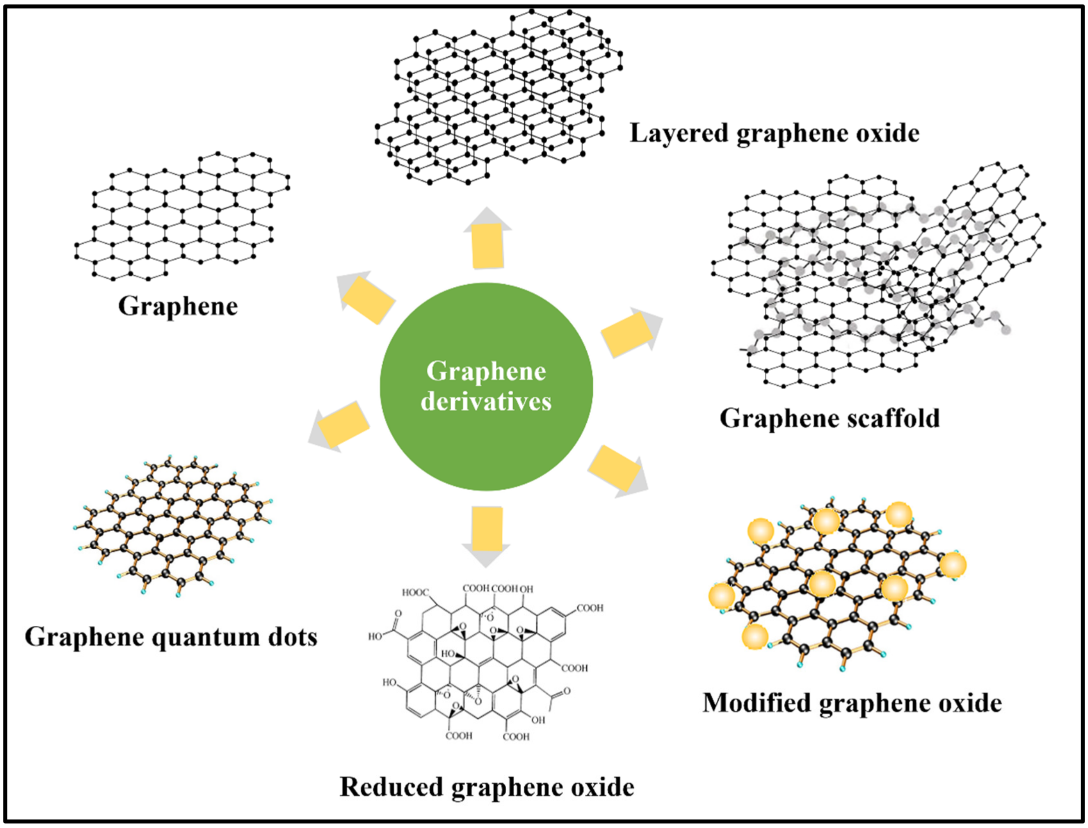

Figure 4. Various forms of graphene and its derivatives.

High-energy oxidants, substances known as plasticizers, adhesives, ignition catalysts, etc. make up the constituent parts of solid propellants, which are a type of composite EM. Since it has a substantial amount of energy and combustible capability, thermite is used as an energetic addition in ammunition and weapons [57]. However, due to its high degree of reactivity, Al oxidizes quickly in the presence of air. As particles aggregate, however, they exhibit poor ignition and combustion qualities, sluggish rate of reaction, decrease in energy discharge, and decreased interaction of reductant with the oxidant. Electrospinning, chemical vapor deposition (CVD), and atomic layer deposition (ALD) have been used to produce core–shell systems, while layer etching and encasing Al with polymer/surfactant have been used to boost the metal's surface responsiveness. In addition to acting as a crosslinker to keep Al in place without increasing becoming unnecessarily bulky, GO can boost mechanical durability and nanoparticle dispersibility [58]. The optimal reaction efficiency with energy production may depend on the total mass proportions of GO, the metal oxide, and Al. By incorporating Bi_2O_3 along with Al into the GO framework via a self-assembly strategy, researchers were able to produce thermite GO/Bi_2O_3/Al in the form of a colloidal suspension stage and, eventually, as a macrostructure exhibiting ultrahigh concentration and high reactivity [58]. Nanocomposite GO/Bi_2O_3/Al was produced by covalent bonding between GO and Al and noncovalent bonding between Bi_2O_3, GO and Al. The amount of energy production increased from 739 to 1421 J/g for the self-assembled GO/Bi_2O_3/Al compared to randomized mixing. Consequently, the thermal efficiency of the thermite was highly affected by how the oxidants and fuel were distributed within it. The direct self-assembly of GO to GO/Bi_2O_3/Al enhanced ignition productivity and decreased susceptibility to electrical discharge [59]. The primary roles that binders made from polymers play in EM are those of fuel, plasticizing agent, curing agent, bonding agent, and ignition catalyst. As a result, the binder ratio has a significant impact on the properties of the propellants. Binders are frequently made of polysulfide rubber (PSR), polyurethane (PU), polyvinyl chloride (PVC), polybutadiene (PBD), and various other compounds [59]. PU's strength, elasticity, and flexibility make up for the material's poor resistance to heat. By using fillers, the thermal endurance and resistance to corrosion can be improved. The thermal endurance as well as structural durability of polymer materials can be improved by adding graphene. The isocyanate units in isophorone diisocyanate (IPDI) can react with the -OH and -COOH of GO sheets. As the resulting IPDI-GO reacts to the -OH at the end of the molecule, the HTPB-GO compound is produced. When IPDI reacted with the -OH remaining in HTPB [60], a graphene + PU composite (MGO + PU) with double crosslinking was formed. The MGO/PU composites outperformed pure PU in terms of CET, tensile reconstruction rate, and stability in dimension. This mixture of IPDI-modified graphene (MGO) and n-butyl acetate displayed a strong dispersal behavior. The tensile strength of 5% MGO + PU was 5.3 percentage points lower compared to that of pure PU, while the 50% elasticity was 31.9% higher. Substituting MGO for inorganic fillers in PU improved the material's resistance to corrosion while decreasing its residual rate. Zhang et al. found that adding Al_2O_3/graphene to PU made it much more durable. The Al_2O_3 surface, with its positive charges, attracted the negatively charged GO by the process of electrostatic adsorption [61]. After that, Al_2O_3/graphene infiltrated the HTPB solution, forming an interlocking structure. Particles of aluminum oxide plus sheets of graphene were placed hierarchically in the matrix of PU after the construction procedure. The final Al_2O_3/graphene/PU composites had superior tensile and thermal characteristics over pure PU in all the domains tested: tensile force, compressing modulus, and heat conductivity. Because it has an inherent strain hardening effect, Al_2O_3 is resistant to distortion and can be fixed by adding sheets of graphene on top of it. Chemical activities, such as electrophilic modification, nucleophilic modification, condensing reaction, and additive reaction, can form covalent bonds involving GO defects as well as functional groups [62]. Because of sp^3 hybridization, GO edges and faces have hydroxyl functional groups. Esters of various kinds can undergo functional modification by reactions with isocyanate or acids such as carboxylic acid. Derivatizing the -OH to create carbamate esters yields isocyanate-treated GO, the degree of functionalization of which is controllable by varying the responsiveness of the isocyanates or the amount of reaction time [62]. Isocyanates can react with the carboxyl group -COOH to produce an amide. Carboxyl groups get located on the surface of GO through sp^2 hybridization. -COOH groups are often utilized to alter a substance's properties by creating an amide/ester through a reaction with -NH_2/-OH. To begin with, inactive intermediates are formed by treating -COOH with acyl halides, which include $SOCl_2$/$SOBr_2$ or carbodiimides [63]. The by-product is subsequently converted into the desired ester or amide by an interaction with either -OH or -NH_2. To attach additional groups onto GO, epoxy-based processes ought to be initially investigated given that sp^3-hybridized epoxy groups prevail on the GO surface relative to -OH and -COOH [63]. Regarding the nucleophilic ring opening interaction involving -NH_2 and the epoxy group, alkalis (which include NaOH, KOH, and NaH) are effective catalysts. Alkali causes -NH_2 to give up an H atom and target the active C, which results in positive -NH_2 bonding to the GO surface. The addition of heteroatom additives [64] to graphene changes the spin distribution as well as charge density of the substance and creates novel sites of activity (substitution including vacancy gaps), allowing the fine-tuning of graphite's mechanical, electronic, electrical, optical, and magnetic characteristics. CVD, hydro- and solvothermal approaches, arc-discharge techniques, thermal processing processes, and others are all common ways for preparing doped graphene [65].

The most effective approach for incorporating heteroatoms into the graphene structure is to search for an extra atom that has an electrical structure similar to C. Nitrogen is frequently used in the creation of heteroatom-doped graphene because of its extreme proximity to carbon. Graphite–nitrogen, pyridinic–nitrogen, and pyrrolic–nitrogen are the three most common coupling topologies for N atoms in nitrogen-doped graphene (NGO). The generated activating region on the material graphene surface can participate directly in catalytic reactions. Ammonia, urea, hydrazine hydrate, and amide are all viable dopants for NGO formation [66]. Researchers have generated an NGO utilizing the hydrothermal method [66], with urea providing the nitrogen supply. Recrystallization in ethyl acetate allowed the formation of NGO composite + CL-20. When compared to undoped graphene, NGO's catalytic efficiency in the thermal decomposition of CL-20 was much higher. The lower temperature and evident activation energy of NGO's exothermic breakdown compared to those of related compounds may be explained by its increasing surface defects along with its active sites. The catalytic decomposition of HMX also provides a little enhancement. Metal catalysts have catalytic active sites on their surfaces; therefore, the particle size of the catalyst has a direct correlation with the amount of catalyst loaded. With regard to catalytic efficiency or selection, a single atom may be superior to a micro, nano, or sub-nano catalyst. Consequently, there has been a lot of interest in doped graphene using single-atom elements such Al and Fe [67]. Theoretical investigations [68] show that the poor physical absorption of certain gas species on pure graphene is replaced by chemisorption. It is possible that this means Al-doped graphene can be used to identify rare gasses. When propellant is burned, these gas molecules are the most common by-products, and propellants rely heavily on Al as their major fuel source. One can expect a huge increase in propellant ignition efficiency if monoatomic-metals-doped graphene is used instead of the standard catalyst [68].

6. Conclusions

Graphene is a brand-new carbon nanomaterial that has exceptional mechanical, optical, and thermal capabilities and a sizable specific surface area. Because of this, it is useful as an additive in energetic materials (EMs). In addition to serving as a catalyst, binder, and desensitizer, graphene improves the mechanical qualities of EMs and ensures security during creation, transportation, and storage. In order to broaden the use of graphene, there has been a lot of interest lately in functionally altering it. Selectively grafting active groups onto the graphene surface is one method that works well to increase the operability of the material. The catalytic activity of graphene is greatly improved by adding energetic groups or molecules. Additionally, adding heteroatom doping modifies the electrical characteristics of graphene, changing the charge distribution and causing activation zones on its surface. Because the use of graphene-based composites to create active composite EMs is a very effective technology, expanding the usability of graphene through modifications is a crucial step. This research also highlights the value of using graphene-based adsorbents to remove hazardous organic and inorganic contaminants from wastewater. These graphene compounds outperform other carbonaceous materials in their ability to effectively remove a variety of contaminants. Even if there are still difficulties in employing graphene-based adsorbents to treat water and wastewater, further study is necessary before commercializing graphene-based materials for actual water and wastewater treatment. New materials made of graphene are proving to be very promising candidates for the treatment of wastewater and water.

Funding: This research did not receive any specific grant from funding agencies in public, commercial or not for profit sectors.

Conflicts of Interest: The authors declare no conflict of interest.

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