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A Comprehensive Review on Graphene Oxide Based Nanocomposites for Wastewater Treatment

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With the paramount development of industry and agriculture sector, levels of different pollutants like, heavy metal ions, pharmaceuticals, organic dyes, biological waste and other pollutants are becoming serious. The ecosystem and human health suffered greatly from the adverse effects of these pollutants. The disposal of these pollutants has become an urgent issue for the human society. Graphene oxide base nanocomposites have generated an excellent extent of focus as desirable alternatives for the adsorptive elimination of contaminants from aqueous systems owing to their enhanced surface area and multiple functional groups for adsorption. Graphene oxide (GO) as a graphene derivative exhibited superior features as obtainable in a graphene sheet. Moreover, the addition of oxygen functional group at the edges and basal plane of graphene further enhanced the efficiency of the graphene by providing sites for the attachment of different metals on the surface. On the underlying adsorption processes, graphene-based nanocomposites for specific contaminants are designed and currently employed for wastewater treatment. This review presents the ongoing development of GO base nanocomposites and their useful applications, understanding how well graphene-based nanocomposites adsorb pollutants and how that relates to the ways in which pollutants interact with adsorbents is crucial. This study highlights newly developed trends in the creation of graphene oxide based nanocomposites to eliminate different heavy metal ions, dyes, pharmaceuticals, and oils spills from effluent water. The focus is on various graphene oxides nanocomposites application for the removal of different pollutants and regeneration of graphene oxide base nanocomposites after several adsorption cycles. Other challenges and potential directions for designing efficient GO based nanocomposites as adsorbents are also presented along with the problems of current studies.

Keywords: Graphene oxide, Heavy metals, Dyes, Pharmaceuticals, Hydrocarbons, Adsorption.

INTRODUCTION

Significant advancement in the industrial sector over the past few decades has caused the rate of processing to rise at the cost of environmental pollution[1]. Wastewater treatment and drinkable water purification are the needs of the time to support the rapid growth of human population and reduce environmental pollution and health risks[2]. Millions of people have experienced health issues because of harmful chemicals polluting the ecosystem and subsequently becoming contaminated. Figure 1 depicts a conceptual illustration of how industries cause water contamination[3].

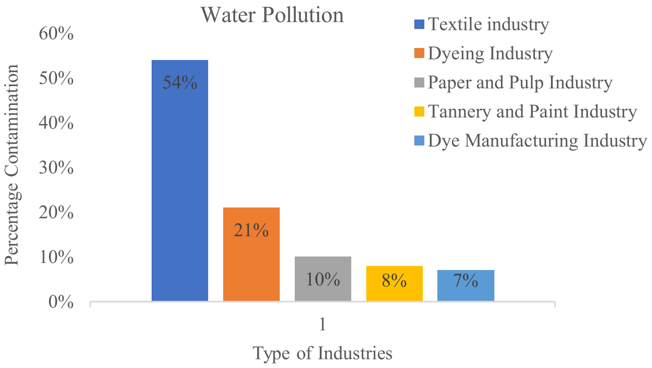

Figure 1. Analysis of the outflow of dye from different sectors

Heavy metal pollution in textile effluent has grown to be a serious environmental issue on a global scale. Coloration is produced by the bathochromic shift caused by the inclusion of metal ions in dye molecules, which produces darker colors. As the textile industry uses a lot of water and because these substances are unable to completely combine with fibers during the dyeing process, a significant amount of wastewater comprising heavy metal ions used for coloration, including Cr, Pb, Cd, and Zn, as well as dyes is added to the freshwater streams without prior treatment[4]. The natural water resources are harmed by these effluents, which have severe negative consequences for the ecosystem and adverse effects on human health. Dyes are mutagenic, and carcinogenic and their exposure can result in allergic reactions like contact rashes, respiratory conditions, eye and respiratory system irritation, and even kidney, bladder, and skin cancer[5]. The extremely harmful dye substances can reduce the passage of light through water, hence lowering the overall purity of water. They can also impact the progress of photosynthesis in the plants present in water and can result in an oxygen shortage in the water increasing the BOD (biological oxygen demand) and COD (chemical oxygen demand)[6].

The term “pharmaceutical wastewater” describes the wastewater produced by pharmaceutical manufacturing processes, pharmaceutical research centers, hospitals, and private residences where medications are utilized or disposed of. Various pharmaceutical substances, such as active pharmaceutical ingredients (APIs), antibiotics, hormones, and other chemicals utilized in the manufacture and consumption of pharmaceutical goods are present in this type of wastewater. Due to the possible negative impact that pharmaceutical chemicals may have on aquatic ecosystems and human health, pharmaceutical wastewater poses a serious environmental concern. These emergent organic contaminants (EOCs), which are extremely difficult for conventional wastewater treatment facilities (WWTPs) to adequately remove from the effluent, end up being released into water bodies. Even at low concentrations, these EOCs can have harmful effects on living things[7]. Along with carbon nanotubes (CNTs) and other nanoparticles (NPs) made of carbon, graphene with a one molecule wide nano-sheets has recently came into focus as an efficient alternative with an array of intriguing significant characteristics, such as the energy conversion, preservation, and catalytic reactions. According to research on water purification and distillation, graphitic carbon strengthens the flow of photoelectrons and greatly improves the photo-conversion efficiency of the system because there are many electrons that are delocalized and connected in the sp^2 geometry of carbon network. Additionally, graphene oxide (GO) demonstrates a high capacity for the absorption of organic compounds in an aqueous environment[8]. In order to eliminate heavy metals from effluent water methods including chemical coagulation, membrane separation, precipitation, electro-kinetics, electroplating, and ion exchange are commonly used[9]. Since GO is a potential nanomaterial with useful characteristics, heavy metal eradication by using materials made of GO can also be taken into consideration. Subsequently, GO can be functionalized with other composites more easily and effectively than graphene. GO is applied for the removal of heavy metals[4] dyes[10] hydrocarbons[11] oils[12] and pharmaceuticals[13] from the wastewater. Adsorption is the most commonly used method to remove the contaminants from the wastewater using GO[14]. Previously reported work on graphene oxide nanocomposites for pollutant removal has demonstrated several key advantages. Firstly, these nanocomposites typically exhibit exceptional adsorption capacity due to the large surface area and abundant functional groups on graphene oxide sheets. This high adsorption capacity allows for the efficient removal of various pollutants, including heavy metals, organic compounds, and dyes, from wastewater. Moreover, the tunable surface chemistry of graphene oxide allows for easy functionalization, enabling the design of tailored nanocomposites for specific pollutant types. This adaptability enhances the selectivity of the material, making it suitable for a wide range of contaminants. Additionally, the potential for nanocomposite regeneration and reuse can contribute to cost-effectiveness and sustainability in wastewater treatment processes. Despite their numerous advantages, previously reported work on graphene oxide nanocomposites for pollutant removal is not without its limitations. One significant challenge is the potential saturation of adsorption sites, particularly when dealing with high concentrations of pollutants. Once these sites are filled, the nanocomposite's efficiency may decrease. Furthermore, the practical scalability of graphene oxide-based nanocomposites remains a concern, as large-scale production can be costly. Additionally, issues related to the release of graphene-based materials into the environment and potential long-term effects require careful consideration. Finally, the stability of nanocomposites in complex wastewater matrices with varying pH, ionic strength, and competing ions can be a challenge, necessitating further research and optimization for real-world applications. In summary, aim of this study is to provide a valuable resource for researchers, engineers, policymakers, and industry professionals by summarizing the current state of knowledge, evaluating the performance of GO base nanocomposite, and guiding future research and development efforts in the field of wastewater treatment for environmental remediation. Figure 2 shows the publications on GO-based composites in recent years. Although publications are relatively limited, it is still a field worthy of development. As can be seen from Fig. 2, the research on GO-based composites shows a gradually increasing trend, indicating that it is attracting more and more attention.

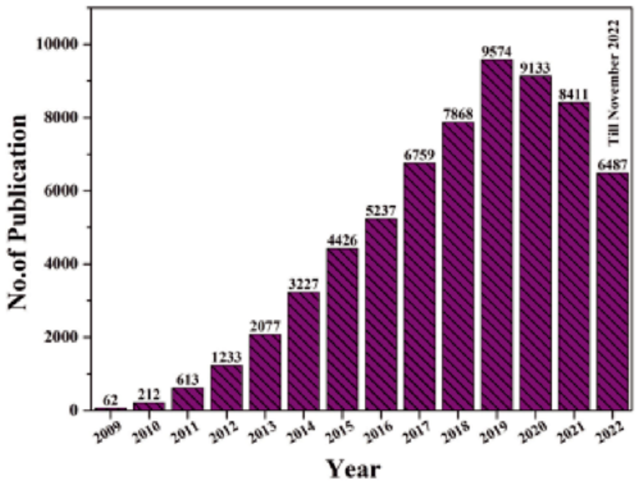

Figure 2. Number of the publications on GO composites

GRAPHENE OXIDE AND ITS PROPERTIES

An altered version of Hummers' method can be utilized to oxidize chemically and disintegrate the graphite flakes to produce graphene oxide(GO)[15], which has epoxy and carbonyl (-CO) groups on its fundamental plane, carboxylic group (-COOH) on the margins, and empty spaces while still retaining some of the pure sp^2 structure of graphite[14]. Graphene and its derivatives are two-dimensional substances with significant surface area as well as outstanding electrical and heat conductivity, and adsorption capability[16]. GO nanosheets turns out to be more hydrophilic than graphene owing to the abundance of functional groups incorporating oxygen (e.g., hydroxyl and carboxylic and epoxide) making it uniformly disperse in clean water and other hydrophilic organic solvents, and readily accessible for interacting chemically with other compounds[17]. The honeycomb lattice of graphene[18] have a distributed system of electrons that is created by the orbital that every carbon molecule in the framework has π orbitals. In all other dimensions, graphene serves as the fundamental building component for all graphitic materials. It can be layered into graphite

(3D), wrapped in graphene nanotubes (1D) or rolled into fullerenes (0D)[6]. Consequently, due to its superior thermal, electrical, mechanical, and optical characteristics, graphene has drawn a lot of interest from researchers, academics, and industries. The exterior surface domain, chemical resistance, resistance to gases, pore size, optical transparency, elastic properties, adjustable bandgap, high mechanical strength and biological compatibility of graphene are also high[19].

In addition to -OH, -OR, -CO, -COOH and other functional groups requiring oxygen[19], the hexagonal carbon arrangement of GO is similar to that of graphene. Beside this easier synthesis, these compounds have several advantages over graphene because of their oxygen-rich groups, such as enhanced soluble capacity and the potential for interface modification, which together have made a wide range of uses for hybrid materials possible. Additionally, GO being analyzed using a range of techniques to create reduced graphene oxide (rGO), which has properties that are comparable to those of pure graphene and reduces the amount of oxygen groups[20]. The tensile force of single layer GO is 207.6 ± 23.4 GPa based on Young's modulus[21]. The sufficient absorbing capacity of hybrid structure and its division of charges in space are made possible by superior electron transporting characteristics, improved catalytic performance, lower density, big surface area, and good mechanical strength of GO. In order to improve photocatalytic activity (PCA), GO also functions as an a powerful electron collector to increase electron transfer by light[22]. In the process of creating GO, disruptions occur in sp^2 covalent orbitals of graphene with the addition of many surface groups, which reduces the electrical conductivity of material and makes GO electrically less conductive. Due to this high impedance, scientists have investigated ways to reduce GO to create reduced graphene oxide(rGO). The electrical conductivity of GO can improved to more than one level of magnitude and can be significantly improved upon reduction, with conduction range varying from $\sim 0.1 - 2.98 \times 10^4$ S m^{-1} [23].

Despite the GO membranes have improved separation capability because of the significant bonding among functional groups and pollutants the penetration of water molecules is hindered by hydrogen bonds that form with molecules of water and the functional groups on GO. Therefore, utilizing membranes that are only dependent on GO makes it challenging to attain an acceptable choice among permeability and selectivity. Since GO nanosheets are strongly electrostatically attracted to one another, GO membranes have an important intrinsic limitation that they are unstable and can break down in aqueous media. The production of a stable membranes of GO with enhanced flux typically requires the addition of molecules for ionic cross-linking, functional group substances, or reducing GO by using chemicals to reduce the functional groups having oxygen[24]. For example, Vacuum filtration was used by Fan et al. to produce the rGO membranes on the PVDF substrate. The hydrothermal reduction duration can be adjusted to alter the extent of reduction of the rGO membranes. The prepared membrane exhibits high-water diffusion and elimination for methylene blue (MB), Congo red (CR), and crystal violet (CV) dyes under the ideal fabrication conditions. As a result, by regulating the hydrothermal time, It is feasible to create the reduced GO membrane with good dye elimination efficiency[25].

Graphene oxide-based nanocomposites

Owing to its superior thermal, mechanical, and optical properties, graphene has drawn attention. Its two major forms are graphene oxide (GO) and reduced GO (r-GO). The traditional way to produce products based on graphene is to first oxidize graphite powder to GO and then reduce it to r-GO[26]. The advanced materials are made by the immense surface areas of graphene and its composites. The process of handling effluent comprising dyes and heavy metal ions has drawn a lot of attention to nanocomposite of GO and r-GO with metal or metal oxide (MO), polymers, SiO_2, sodium alumino silicates, and other inorganic materials, including metal organic framework (MOFs). These hybrid materials can be used as photo-catalytic or adsorbent agents[27, 28]. The GO/r-GO composites show decreased gaps in their energy levels and reduced frequencies of electron-hole pairs, which results in superb radiation adsorption and improved photo-degradation potential[22]. As can be seen from the adsorption capabilities, the nanocomposites made of GO has been extensively studied for the adsorption of dyes and pharmaceuticals. Some research has been collected to promote the utilization of materials made on GO as adsorbents and demonstrate their effectiveness in order to gain insight into batch processes. Biopolymers in conjunction with graphene materials have also been studied due to the growing demand for green chemistry. The adsorption of organic molecules is perfect for the employing of graphene materials due to their biocompatibility, biodegradability, availability, high reactivity, and ease of separation[29].

Singh et al used the solvothermal method to create ornamented ZnO NPs on GO NP. The Cr(VI) adsorption kinetics of prepared nanocomposites and their elimination capability are enhanced by the coating of ZnO NPs on graphene oxide(ZnO-GO). The efficiency of removal decreases under alkaline circumstances while increasing from a high acidic to a neutral pH. The created nanocomposites show great promise for removing Cr(VI) from natural wastewater networks and water bodies[30]. Kaur et al used simple sonication to create graphene oxide (GO) and $MgFe_2O_4$ nanoparticles (NPs). Its potential for removing the particles Ni(II) and Pb(II) from water was examined. Ni(II) and Pb(II) ions each have a maximal adsorption potential of 100.0 mg/g and 143.0 mg/g, respectively. Along with having a greater capacity for adsorption, magnetic characteristics of nanofabricated composites make it possible to reuse nanocomposite incorporating metals for adsorption and release of pollutants[31]. Choi et al prepared a mixture of chitosan (CS), graphene oxide (GO), and gadolinium oxide (CGO-Gd) to remove As(V) from water through adsorption. At pH 6.0 and for concentrations of 0.1 and 0.3 g/L of prepared nanocomposite, respectively, the elimination capacities of As(V) were 252.12 and 128.20 mg/g. Additionally, the CGO-Gd can be used to remove As(V) up to five times, proving the material's viability and affordability[32]. Marjani et al prepared samples of GO were created by the Hummer method and used to create composite membranes. Water was used as a non-solvent in the wet phase inversion method to create PES NC membranes. Purified water flow, salt elimination, dye preservation, and elimination of heavy metals were evaluated in order to gauge the effectiveness of the prepared membranes that had been created and can be used in cleaning of polluted water[1, 33].

Synthesis Methods of GO

Graphene oxide can be made by various methods, each of which has benefits and restrictions of its own. Although appealing for creating high-quality GO, mechanical exfoliation[34] has limitations in relation to the viability of fabrication on pilot scale. Low yields have been observed for both epitaxial growths on silicon carbide (SiC) wafer techniques and mechanical exfoliation (by ultrasonication or stirring). British chemist B. C. Brodie made the initial effort to fabricate graphite oxide in 1859 while researching the reactivity of flake graphite. This method uses $KClO_3$ as an oxidizing agent and is known as the chlorate pathway. Staudenmaier[35] made improvements to the procedure in 1898 by using concentrated sulfuric acid in addition to boiling nitric acid. With this change, the method was made simpler while the quantity of produced oxidized graphene oxide increased[36].

Graphite oxide can be created by oxidizing graphite, and then this graphite oxide can be exfoliated to create GO. The technique of synthesis, which affects the number and kind of groups having oxygen in the formed GO, has an immense impact on the properties of the substance[37]. The Hummers method developed by Hummers and Offeman, is the most popular and efficient approach ever. The conventional technique for the synthesis of GO is comparatively quick. This process involves adding excessive potassium permanganate, sulfuric acid, and very little sodium nitrate to the reaction combination. The reaction period varies from 8 to 12 hours. Due to the fact that this method prevents the production of explosive ClO_2, it is much safer[38]. At the end of the process, a dilute solution of H_2O_2 is used to neutralize the excess potassium permanganate. The creation of potentially explosive gases was prevented in this process. Ice baths were used to closely regulate the temperature, ensuring that it stayed as low as possible throughout the reaction[37]. Figure 3 shows the chemical oxidation processes used by Brodie, Staudenmaier, and Hummer to create graphene oxide.

Synthesis Method of GO Nanomaterials

There are different techniques to produce graphene oxide nanomaterials. Some of them are explained briefly:

Hydrothermal Technique

This method involves creating nanoparticles in a liquid medium at high pressure and temperature[40]. Many scholars have looked into using the hydrothermal method to prepare GO nanostructured materials[41]. Organic molecules in alkaline environments are the constituents of the hydrothermal process. Despite being regarded as cost-effective and environmentally friendly, hydrothermal technology typically entails high temperatures. The autoclave's temperature range varies between 160 and 180 °C. There are some restrictions, though, including the inability to observe the crystal material's growth in the autoclave and the high expense of the apparatus[20, 42].

Solvothermal Technique

This method uses non-aqueous fluids at high pressure and temperature to prepare various nanoparticles. Two types of solvothermal processes can be distinguished: production in basic environments and in with the inclusion of organic molecule precursors[43]. There are many benefits to using a solvothermal method to make graphene oxide, including the fact that it is safe, economical, and produces almost no byproducts during the reaction[44].

Co-precipitation Technique

Co-precipitation of metal cations from $C_2O_4^{-2}$, CO_3^{-2}, $C_6H_5O_7^{-3}$, and OH^-, among other materials, is involved in this. At an appropriate temperature, these precipitates are transformed into granules. The use of unfavorable impurities that also co-precipitate with the analyte is one drawback of this technique. By re-precipitating the analyte, which causes inclusion and occlusion, this flaw can be fixed[45, 46]. Figure 4 shows the different methods for the GO synthesis.

Sol-gel Technique

This is a straightforward and cost-effective wet-chemical technique used to create composite materials with outstanding control over size. In this method, the solution (sol) progressively develops into gel-like material that is composed of both a solid and a liquid phase. Aqueous and non-aqueous sol-gel synthesis are the two types of sol-gel methods[47]. The first stage in creating a rational

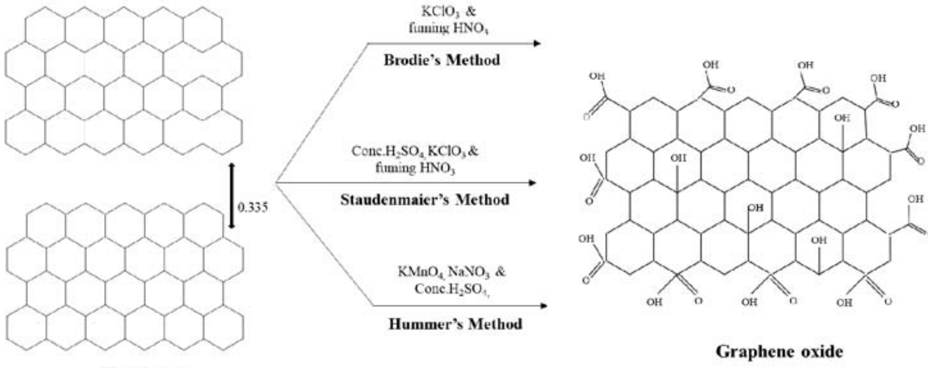

Figure 3. Diagrammatic representation of the chemical oxidation processes used by Brodie, Staudenmaier, and Hummer to create graphene oxide[36, 39]

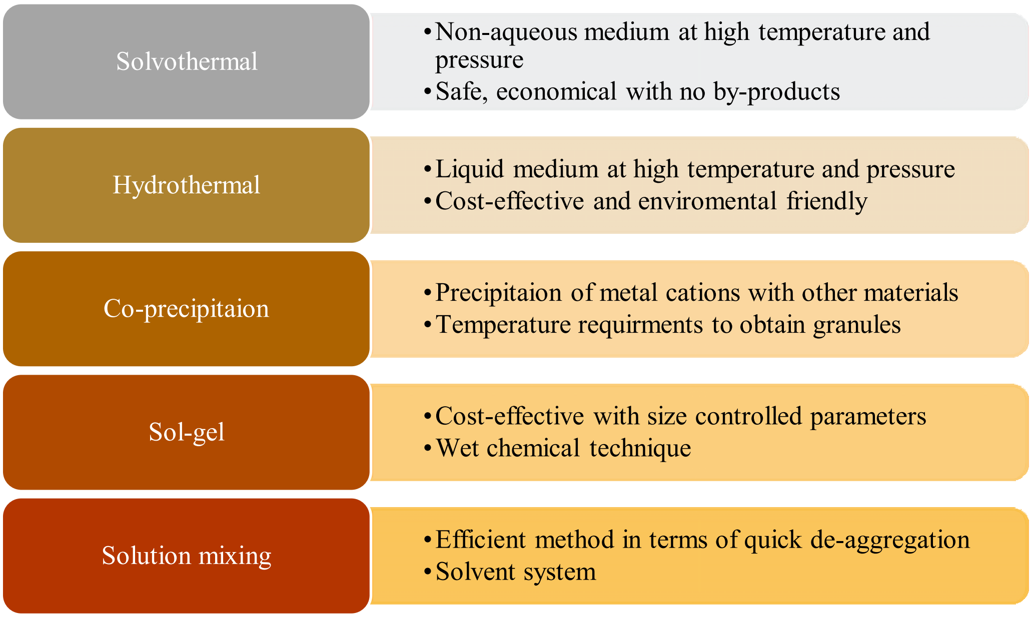

Figure 4. Different GO Synthesis Methods with some properties

synthesis for non-aqueous sol-gel preparation of metal oxide (MO) NPs is to elaborate the chemical formation mechanism alongside studies on the crystallization process. Conversely, to guarantee that this method yields comprehensive results, it is necessary to investigate various characterizations, including microscopy and crystallography, among others. It has been found that using common methods like NMR and GC-MS, among others, allows for simple to investigate the changes that occur in organic entities in a chemical reaction[48].

Solution Mixing Technique

This method is frequently used to create GO/metal oxide nanomaterials because of its modest temperature, quick de-aggregation, and equal reinforcement distribution of the composite it produces. The fundamental method for combining solutions is in a solvent system. This method uses electrospinning to combine two distinct nanoparticles in a solution. The primary drawback, however, is the possibility for extra material to leak because there is no chemical bond between it and the base[49]. Table 1 shows some of the method used for the synthesis of GO nanocomposites.

FACTORS AFFECTING THE REMOVAL OF POLLUTANTS

The use of graphene oxide (GO) based nanocomposites for wastewater treatment is a promising approach due to the unique properties of graphene oxide, such as its high surface area, strong adsorption capacity, and the ability to functionalize its surface. Several factors can affect the removal of pollutants using graphene oxide-based nanocomposites for wastewater treatment[48]. The properties of graphene oxide, including its surface area, functional groups, and sheet size, can influence its adsorption capacity and reactivity towards pollutants. i) the choice of materials used in the nanocomposite, such as other nanoparticles or polymers, can greatly affect the composite's pollutant removal efficiency. ii) the pH of the wastewater can significantly impact the removal of pollutants. The surface charge of graphene oxide can change with pH, affecting its adsorption capacity for different types of pollutants. iii) the duration of contact between the nanocomposite and the wastewater is important. Longer contact times often lead to better pollutant removal, but this may not always be feasible in practical applications. iv) temperature can influence the adsorption and desorption kinetics. Higher temperatures can generally enhance the removal of pollutants due to increased molecular mobility. v) the initial concentration of pollutants in the wastewater can affect the removal efficiency. At higher initial pollutant concentrations, the nanocomposite may become saturated more quickly. vi) The amount of graphene oxide-based nanocomposite added to the wastewater is crucial. Using an appropriate dose is essential to ensure efficient removal while avoiding unnecessary waste. vii) the composition of the wastewater, including its chemical and physical properties, can vary widely. Different types of pollutants and water matrices may require specific considerations for optimal removal. viii) the ability to regenerate and reuse the graphene oxide-based nanocomposite can be important for cost-effectiveness and sustainability[38]. It's important to note that the effectiveness of graphene oxide-based nanocomposites can vary depending on the specific pollutants and wastewater characteristics. Optimization and customization of these factors are often required for each wastewater treatment application.

METHODS FOR POLLUTANT REMOVAL

The water bodies are heavily contaminated by environmental contaminants like dyes, organic pollutants, and heavy metals. However, because of their high cost and poor economic viability, these techniques are not widely employed. The most adaptable and frequently used method of water treatment is adsorption[3]. Some other methods for pollutant removal are ion exchange, chemical precipitation, coagulation, bioremediation, and membrane filtration. The adsorption procedure, among the methods mentioned above, is a practical way to treat discharge from factories wastewater having low running expenses, few pollution problems, and the

Table 1. Different GO nanocomposite with their synthesis methods

Nanocomposite	Synthesis Method	Adsorption Capacity mg/g	Type of pollutant removed	References
PCN-222/GO-COOH	Solvothermal technique	426	U(VI)	53
GO–MnO_2	Hdrothermal	149.253 178.253	MO MB	54
GO-$MnFe_2O_4$	Hydrothermal	621.11	Pb(II)	55
CTS/PAA/GO	Sol-gel	296.5 280.3	MB FY3	56
SGA/ODA/GO	Sol-gel	52–178 g/g	Oil	57
GO/Fe_3O_4	coprecipitation	279.62	Cr(III)	58
Ag/rGO	coprecipitation	–	Dyes	59
rGO–ZnS	Facile one-step chemical approach	27.54 20.04	MG EV	60
MCSGO nanocomposite	ultrasound agitation	66.15 112.6	Safranin O (SAF) Indigo carmine	61
CdO-GO nanocomposites	electrochemical exfoliation	430	Cr (VI)	62

most economical method to take heavy metals out of the industrial waste. Adsorption is the process by which a liquid material binds to a solid surface. The reversible desorption method used in this adsorption procedure allows for the regeneration of absorbents, which can then be used repeatedly[50]. Adsorption is a prominent technique in the industrial water discharge treatment methods owing to its easier use, flexible to handle, and has a small footprint. The effectiveness of the adsorption technique is influenced by the adsorbent's polarity, appropriate functionalities or functional groups, pore volume or pore size distribution and increased surface area[51]. Adsorbent dosage, initial concentration, and temperature are just a few of the variables that affect how well effluent pollutants are eliminated[52].

APPLICATIONS OF GO IN WASTEWATER TREATMENTS

Lately, graphene and its functionalized composite materials are effectively utilized for wastewater treatment. Graphene and its modified substances such as GO are highly effective for eliminating hazardous heavy metals from polluted water because of their substantial surface area, a significant amount of intrinsic functional groups. In addition, GO can provide wastewater treatment technology at a lower expense than other approaches. As a result, it can be used to treat wastewater that contains organic effluents more than allowed levels. Since GO can be employed to eliminate all kinds of impurities from wastewater, it can be an effective alternative for activated carbon-based water filters. The outcomes are very effective at eliminating metal ions, organic pollutants, and PAHs among other toxins[3, 63]. The inclusion of graphene oxide (GO) in wastewater treatment systems has shown promising effects on improving efficiency. However, the specific impact of GO on wastewater treatment efficiency can vary depending on the specific application and context. Here are a few potential effects of including GO in wastewater treatment systems: *Enhanced adsorption capacity:* Graphene oxide has a large surface area and high adsorption capacity, making it effective for removing various pollutants from wastewater. Its presence in the system can enhance the adsorption process, leading to more efficient removal of contaminants[64]. *Improved membrane performance:* Graphene oxide can be incorporated into membranes used in filtration processes. The addition of GO can enhance the membrane's selectivity, permeability, and fouling resistance, resulting in improved separation efficiency during wastewater treatment[65]. *Catalytic activity:* Graphene oxide-based materials exhibit high catalytic properties, allowing them to facilitate the degradation of organic pollutants through advanced oxidation processes. The breakdown of contaminants can be increased by adding GO-based catalysts to wastewater treatment systems, increasing treatment effectiveness [66].

Removal of Dyes

Organic dyes are introduced to the water bodies by petroleum leaks, agricultural waste, and industrial waste. Regulatory agencies and communities around the world have been very concerned about the acute harmful effects, environmental safety, and perseverance for a long time. Lots of water is needed for the dyeing process, and water is continually discharged into the environment. The aquatic ecosystem is destroyed as a result of the wastewater from the dyeing industries, which contains dangerous and cancer-causing dyes which are harmful for fish, marine animals, and mammals as well as pose significant risks to aquatic plants' ability to photosynthesize[10]. GO, which is synthesized by chemically altering graphene to give it functional groups that contain oxygen and a high surface area, can be a powerful adsorbent for dye adsorption[67]. Ikram et al prepared reduced graphene oxide (rGO) using a heat-treatment technique, and GO was synthesized by a modified Hummers process. By utilizing a hydrothermal technique, various concentrations of silver were incorporated into the nanosheets of GO. Ag/rGO induces photocatalytic activity, which destroys the MB content. These results imply that produced nano catalyst exhibits no hazardous behavior during water treatment and works superbly to eliminate various toxins from effluent water[8]. Das et al prepared GO-CS-PVA hydrogel nano polymer to eliminate CR dye from a mixture. The adsorption of dye onto the polymer made from GO was done in a batch setup. By examining FTIR and SEM data, prepared Graphene oxide reinforced hydrogel polymer (GORHP) was described. The efficiency of dye adsorption for 20 mg/L of dye solution at pH 2 was determined to be 88.17% [67]. Khadim et al examined the removal of methylene blue (MB) and rhodamine B (RhB), from wastewater using the synthesized GO. SEM were used to describe the surface shape of the adsorbents. The studies show the hydrophilic character of the GO. The migration of GO to the membrane surface will increase the membrane's hydrophilicity since it contains hydrophilic

Table 2. Different GO nanocomposites for the removal of dyes

GO Nanomaterials	Dye Removed	Adsorption Capacity (mg/g)	Removal Efficiency	Degradation Time	References
Ag/rGO	MB		100	120 min	[8]
GO/Chitosan–PVA polymer	CR	–	88.17	90 min	[67]
TiO_2-NW-GO	MB	–	98	120 min	[16]
GO/TiO_2 composite	MB	–	99	4 h	[69]
GO/MXene membrane	NR, MB, CV, BB	–	99.5	1 h	[24]
F-GO	MB	403	91.2	1 h	[70]
IC-rGO	RhB	686	10.3	2 h	
$BaTiO_3$/GO composite	MB	–	95	3 h	[71]
MMGO composite	MV	243	96	2 h	[72]
	AR88	303	73		

functional groups. The highest adsorption capability of MB and RhB on GO was determined to be 403.3 and 686.6 mg/g, respectively[68]. Adsorption of dye onto the polymer made from GO was done in a batch setup. By examining FTIR and SEM data, prepared GORHP was described. The efficiency of dye adsorption for 20 mg/L of dye solution at pH 2 was determined to be 88.17%[67]. Table 2 represents some GO nanomaterials used for the removal of dyes.

Removal of Heavy Metals

Two different electrostatic and Van der Waals interactions are considered responsible for removing metal ions from water-based systems. The charged surfaces generated on the absorbent surface correspond to the electrostatic interaction, and the combination of functional groups with metal ions is correlated to the van der Waals interaction. The -COOH group are located on the corners of the highly disrupted graphite carbon network with sp^2 hybridization in GO. The coordination of the metal ions is made possible by the presence of -OH and -COOH in GO. Metal ions engage in competition with one another to produce these interactions, and they function differently when interacting with nanocomposites. Metal ions are eventually eliminated through a 3-step process that involves external metal ion diffusion, diffusion into the nanocomposite, and absorption[1]. Farooq et al used an easy technique for making magnetic GO (MGO) which involved synthesizing the GO surface with Fe(III). MGO was characterized using UV-VIS, FTIR, SEM, XRD, and VSM. Using Atomic Absorption Spectroscopy, the effective removal of harmful heavy metal ions was investigated. Pb(II), Cr(III), Cu(II), Zn(II), and Ni(II) ions had the greatest adsorption capacities, with values of 200, 24.330, 62.893, 63.694, and 51.020 mg/g, respectively[73]. Le et al created a hybrid adsorbent called MGOCS, which is made of magnetite, chitosan, and graphene oxide. For RB19 and Ni (II), the highest adsorption capability of prepared composite was 102.06 and 80.48 mg/g, respectively. The experimental findings showed that the beads' high capacity for adsorption, ease of recovery, and ability to be reused make them potentially useful as an extremely operative and environmentally favorable adsorbent to eradicate organic dyes and heavy metals from water[74]. Table 3 represents some of the GO nanocomposites used for the removal of heavy metals. Modi et al created ZIF-67/rGO to improve the removal efficiency of PES-HFMs. The results showed that lead has an absorption capacity of 86.4 mg/g and copper has a capacity of 66.4 mg/g. The nanocomposite HFMs could be readily renewed and used for an additional five filtration cycles[75]. Wu et al prepared and used CMC/SA/GO-Fe_3O_4 and removed Cu (II), Cd(II), and Pb(II) from the polluted water. Due to the addition of GO/ Fe_3O_4, the prepared magnetic gel beads displayed greater stability and could readily separate from the wastewater. Cu (II), Cd (II), and Pb(II) were determined to have maximum adsorption capacities of 55.96, 86.28, and 189.04 mg/g, respectively. After 5 rounds, the prepared beads still retained 90% of the adsorption rates[76].

Removal of Pharmaceuticals

The use of numerous new pollutants has grown because of modern lifestyle and related economic growth. These pollutants are organic compounds, such as polymers, additives, beauty products, detergents, and medicated items. Pharmaceuticals and personal care products (PPCPs) have gained attention from these pollutants owing to their importance in daily living, extensive fabrication, and high demand[77]. Long-term bioaccumulation of PPCPs on the environment's flora and fauna can result in antibiotic resistance as well as serve as carcinogenic agents[78]. Due to its cheap expense, benign operating conditions, low energy consumption, and other factors, adsorption with a solid material may be the best option in this situation for handling PPCPs in a water setting[79]. Activated carbon (AC)[80], clays[79], graphene-based materials[81], metal nanoparticles[82], and metalorganic frameworks (MOFs)[83] are just a few of the many solid sorbents that has been tried and even used at the pilot scale. Nanocomposites based on graphene oxide have demonstrated promise in the elimination of active pharmaceutical ingredients (APIs). The removal of APIs using graphene oxide-based nanocomposites has been explored in different applications like, *removal of APIs from water:* Graphene oxide–agar–agar hydrogel has been prepared and used for the efficient removal of methyl orange dye from water[84]. The hydrogel exhibited adsorption properties that allowed for the effective removal of the dye. *Hemostatic materials:* In the field of hemostasis, the use of graphene oxide-based nanocomposites has been investigated. For example, hydrophilic chitosan/graphene oxide composite sponges have been developed as hemostatic materials. These materials promote blood clot formation and can be stripped off without causing secondary bleeding or infection risks[85].

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Table 3. GO nanocomposites and type of heavy metal removed

Graphene Oxide Nanomaterial	Heavy metal Removed	PH	Maximum Adsorption Capacity (mg/g)	Removal Efficiency	Reference
MGO	Pb	5.0	200	99.972 %	73
	Cr	6	24.330	97.783 %	
	Cu	6	62.893	96.651 %	
	Zn	7	63.694	91.883 %	
	Ni	8	51.020	95.283 %	
MGOCS	Ni	6	80.48	62%	74
ZIF-67/cGO	Pb	–	86.4	95%	75
	Cu		66.4		
GONRs	As	6	155.61	–	86
	Hg		33.02		
PAO/GO	Pb	5	116.7	–	87
	Ag		258.6		
	Cu		192.2		
	Fe		167.9		
GO/CA	Pb	5	602	99.6%	88
	Hg		374	99.2%	
	Cd		181	98%	
SA-PAM/GO	Cu	5	68.76	80%	89
	Pb	5.5	240.69	60%	
ZnO-GO	Cu	4	33.5	92.9%	90
	Al		19.9	95.6%	
GO-SiO_2	Pb	–	527	–	91
	As		30	–	
TFN/FO/GO	Pb		7.82	99.9%	92
	Cd		6.99	99.7%	
	Cr		1.88	98.3%	
CS-GO	Cr	2.0	104.16	96%	93
GO/Fe_3O_4	Cr	–	–	52%	94
Fe_3O_4-GO/SiO_2	Cr	–	182.98	94%	95
	Co		116.35	96%	
	Ni		226.08	95%	
	Cu		149.59	92%	
	Cd		100.81	91%	
	Pb		168.55	84%	
	Ag		141.09	82%	
GO-$MnFe_2O_4$	Pb	–	621.11	74%	55

ditives, beauty products, detergents, and medicated items. Pharmaceuticals and personal care products (PPCPs) have gained attention from these pollutants owing to their importance in daily living, extensive fabrication, and high demand[77]. Long-term bioaccumulation of PPCPs on the environment's flora and fauna can result in antibiotic resistance as well as serve as carcinogenic agents. As a result, there is an increasing need to completely remove these PPCPs from water and effluent[78]. Due to its cheap expense, benign operating conditions, low energy consumption, and other factors, adsorption with a solid material may be the best option in this situation for handling PPCPs in a water setting[79]. Activated carbon (AC)[80], clays[79], graphene-based materials[81], metal nanoparticles[82], and metalorganic frameworks (MOFs)[83] are just a few of the many solid sorbents that has been tried and even used at the pilot scale. Nanocomposites based on graphene oxide have demonstrated promise in the elimination of active pharmaceutical ingredients (APIs). The removal of APIs using graphene oxide-based nanocomposites has been explored in different applications. Here are a few examples: *Removal of APIs from water:* Graphene oxide–agar–agar hydrogel has been prepared and used for the efficient removal of methyl orange dye from water[84]. The hydrogel exhibited adsorption properties that allowed for the effective removal of the dye. *Hemostatic materials:* In the field of hemostasis, the use of graphene oxide-based nanocomposites has been investigated. For example, hydrophilic chitosan/graphene oxide composite sponges have been developed as hemostatic materials. These materials promote blood clot formation and can be stripped off without causing secondary bleeding or infection risks[85].

In-depth research was made by Delhiraja et al on the sorption of drugs using a composite adsorbent made of graphene oxide (GO). Activated carbon and chitosan had been incorporated, according to structural analysis, onto the GO. The adsorption kinetics for both groups of pollutants supported a pseudo-second-order kinetic model. As opposed to the Freundlich isotherm, the Langmuir isotherm demonstrated a superior fit. ACP, CBZ, BPA, CAFF, and TCS were found to have maximum adsorption capabilities of 13.7, 11.2, 13.2, 14.8, and 14.5 mg/g, respectively[96]. Liu et al prepared a composite made of GC/MGO-SO_3H and used as an absorbent for pollutants in the atmosphere. The GC/MGO-SO_3H behaves in a superparamagnetic manner as is usual. Through batch tests, the prepared composite's adsorption properties to pharmaceuticals were examined. Ibuprofen and tetracycline, respectively, have higher maximal adsorption capacities upon increasing the temperature from 298 to 313 K were 113.27 to 138.16 mg/g and 473.25 to 556.28 mg/g, respectively[97]. Balasubramani et al investigated the adsorption-mediated elimination of metformin from wastewater sample using GO. It was discovered that metformin had a maximal adsorption capacity of 122.61 mg/g. The data match the pseudo-first-order model well, according to the kinetics. According to desorption experiments, the GO was regenerated for six cycles using a 1 N NaOH solution[98]. Some of the GO base nanomaterials for the removal of pharmaceuticals are covered in Table 4.

Removal of Oil/ Hydrocarbons

Industrial wastewater treatment has elevated to the top of the priority list due to the long-standing worldwide issues of shortages of water and sustainable water use. While traditional methods are not enough for purifying produced water in order to meet the new rules, refineries may get rid effluent water in many ways, including, in the situation of remote areas, dumping it in the water following purification to adhere to strict regulations pertaining to the environment[99]. In cracks of the reservoir, oil droplets may aggregate with scale and sand, lowering permeability[100]. The cost of well interventions is increased by both scale growth and greasy water[101]. Due to the growing need for environmental and ecological protection, the cleanup of oil spills in water and the cleaning of wastewater that contains oil have received a lot of attention. Oils can currently be removed from water networks using techniques like in-situ burning, skimming, bioremediation, chemical dispersion, and adsorption[102].

Graphene oxide can be used to remove oil droplets and reduce growing the levels of ions to suitable values, which will fix this issue. The benefits of using GO in produced water treatment include superior permeation, reduced space requirements, simple operation, and the lack of chemical input. Water can be efficiently cleaned of oils by using a graphene-based adsorbent with hydrophobic and oleophilic characteristics[12]. Javadian et al used oleic acid (OA) to change the hydrophobicity characteristics of Fe_3O_4 nanoparticles to create Fe_3O_4-OA. After that, new superhydrophobic nanosheets of Fe_3O_4-OA/GO were created by functionalizing GO nanosheets with prepared nanocomposite. To eliminate oil from oily effluent, a prepared synthetic nanocomposite was used as a quick, easy to be recycled, efficient performance, economical, and eco-friendly nano emulsifier. Testing for demulsification showed that the prepared nanocomposite had outstanding demulsification performance (99.99%) and the capacity to demolish an water/oil mixture six times[108]. Ferrero et al synthesized rGO-coated microfibers (GCMs). The resulting GCMs exhibit high oleophilicity and hydrophobicity, which is suitable for recovering crude gasoline from mixtures of oil and water. After 10 adsorption-desorption cycles, the GCMs' maximum uptake capability is 6.78 g/g, with a reduction of 13.5%. After a single application of GCMs, removals of 97.90% and 87.58% of petroleum oil were made[109]. Other GO used for the removal of hydrocarbons are given in Table 5.

MECHANISM OF REMOVAL

Adsorption techniques using graphene oxide-based nanocomposites have shown promise in the removal of dyes, medicines, and hydrocarbons. Because of their high surface area, adjustable characteristics, and great adsorption capacity, these nanocomposites are useful in the treatment of wastewater and environmental remediation. Several aspects contribute to the adsorption mechanism of graphene oxide-based nanocomposites. For instance, the enormous surface area of graphene oxide gives multiple places for adsorption interactions with the target pollutants. The oxygen-containing functional groups on the surface of graphene oxide contribute to its adsorption capability [110]. These functional groups can form hydrogen bonds, electrostatic interactions, and π-π stacking interactions with the pollutants, facilitating their

Table 4. Different GO nanomaterials for the removal of pharmaceutical

GO Nanomaterial	Type of Pharmaceutical removed	Adsorption Capacity (mg/g)	Type of interaction	References
GO-AC-CS	ACP, CBZ BPA CAFF TCS	13.7 11.2 13.2 14.8 14.5	Hydrogen bonding Electrostatic and π−π interactions	96
GC/MGO-SO_3H	Ibuprofen Tetracycline	138.16 556.28	Electrostatic interaction	97
GO	Metformin	122.61	Vander Waals forces	98
GO–IL	SMZ CBZ KET	99.155 87.875 61.875	Hydrophobic interactions	103
GO	Diclofenac	653.91	Hydrogen bonding Hydrophobic attraction, π−π interactions	104
mGO-Si	sulfamethoxazole	15.46	Hydrogen bonding π−π interactions	105
GO	Norfloxacin	374.9	Vander Waals forces	106
β-CD/rGO	Naproxen	361.85	Hydrogen bonding	107

Table 5. Some GO nanomaterials/nanomaterials used for the removal hydrocarbon

GO Nanomaterials	Hydrocarbon Removed	Maximum Degradation rate	Maximum Adsorption Capacity (wt/wt)	References
GO	n-alkane BTEX	70% 77.8%	–	113
CNF/GOAs	Oils and organic solvents	–	20–286	114
PVA-co-PE nanofibrous aerogels	Oils and organic solvents	–	25–53.29	115
Melamine sponge -ZIF-8	Various oils	–	10–38	116
PVDF/SiO_2/GO	Crude oil	99.65%	129–264	117
a-GPCCA	Oils	88.8%	155–288	118
CNF/PVA/GO carbon aerogels	Various oils	–	97	119
CNF/GO	Various oils	100%	39–68	120
NR/rGO	Crude oil	70%	17.04	121

removal from the water[7]. In the case of dyes, the π-π stacking interactions between the conjugated systems of the dye molecules and the graphene oxide sheets play a crucial role in their adsorption[111]. The presence of aromatic rings in many dyes allows for favorable interactions with the graphene oxide surface. Additionally, electrostatic attractions between the charged functional groups of the dyes and the charged surface of graphene oxide also contribute to their adsorption[112]. The presence of polar functional groups such as amines, hydroxyls, and carboxyls on medicinal substances allows hydrogen bonding with the graphene oxide surface, resulting in adsorption[7]. The hydrophobic interactions between the pharmaceutical compounds' hydrophobic portions and the graphene oxide sheets improve the adsorption process[111].

In the case of hydrocarbons, the - interactions between graphene oxide sheets and hydrophobic hydrocarbon molecules are important in their adsorption[111]. The hydrophobic properties of graphene oxide, as well as its capacity to form clustered structures, make it an ideal substrate for hydrocarbon adsorption. Adsorption of hydrocarbons onto the graphene oxide surface can be improved further by using other functional elements, such as iron oxide nanoparticles, which promote adsorption capacity and give magnetic separation capabilities[122]. Depending on the degree of contamination (heavy-metal ion concentration), different techniques are used for eliminating heavy metal from water. Chemical precipitation is efficient, with a removal rate of up to 99%, when the initial contamination level is significant, as in industrial wastewater. The subsequent pollution produced by this technique, however, is sludge with mixed pollutants, which requires additional extraction or cleaning. This not only increases treatment intricacy but also lowers the worth of any heavy metals retrieved from the wastewater. On the contrary, chemical precipitation is impractical when the pollution concentration is minimal, such as in point-of-use water, because of the significant input of precipitation chemicals[123].

Adsorption and ion exchange are two techniques for removing traces of heavy metal contamination from water. Nanomaterials with a large surface area have been studied for the adsorption of heavy metal and its elimination, and the elimination rate may exceed 90%. Robust attaching ions predominate active sites of the surface and significantly decrease the adsorption of heavy metal ions with poor binding because when several ions mix in the water that is polluted, the degree of binding between the surface sites of sorbents and heavy-metal ions determines the extent of adsorption. This makes it challenging to treat numerous ions from contaminated water at once. Additionally, some research has been done on the electrochemical deionization technique for removing heavy metal particles from water. Through a redox-based pseudo capacitor mechanism, the potential of heavy-metal reduction can be increased more than pure adsorption, and kinetics is quicker than pure adsorption. Nevertheless, it is still difficult to separate various heavy-metal atoms[124, 125]. The development of highly effective nanomaterials for the pollutant's removal, an understanding of the contact process between adsorbents and heavy metal ions is essential. The elimination of heavy metal ions at solid-liquid boundaries may be significantly influenced by a variety of interaction processes, including ion exchange, surface complexation, and precipitation. Multiple experiments at the macroscale, however, fail to offer a complete grasp of the microscopic nature of the interaction mechanism. In order to thoroughly study the reaction mechanism, spectroscopic methods such as FT-IR, XPS, and XAFS are suggested and used[126]. With exceptional spatial precision at the molecular level, FT-IR and XPS spectroscopy are widely used surface chemical analysis techniques that can provide structural details about materials[127]. The functional groups which are present on the GO adsorbents' surface that serve as the primary regions for surface complexation and electrostatic interactions, are primarily responsible for the heavy metal adsorption. The adsorption performance will be greatly impacted because the interactions caused by π electrons are strongly affected by the pH and ionic strength of the solution. Additionally, through cation-/anion-interactions, the scattered electrons on the graphene substrates can interact with both anions and cations of heavy metal as shown in Figure 5. Electrostatic, and hydrogen bonding interactions take place among dyes and adsorbents made of graphene emphasizing the role of the sp^2 carbon atoms and surface functional groups of graphene[63].

RECOVERY OF GO AFTER WATER TREATMENT

Regenerating adsorbents after adsorption reduces the need to dispose of used adsorbents, making it an important factor in practical uses. Depending on the charge signatures of the heavy metal ions, different contaminants may be utilized to get rid of them from

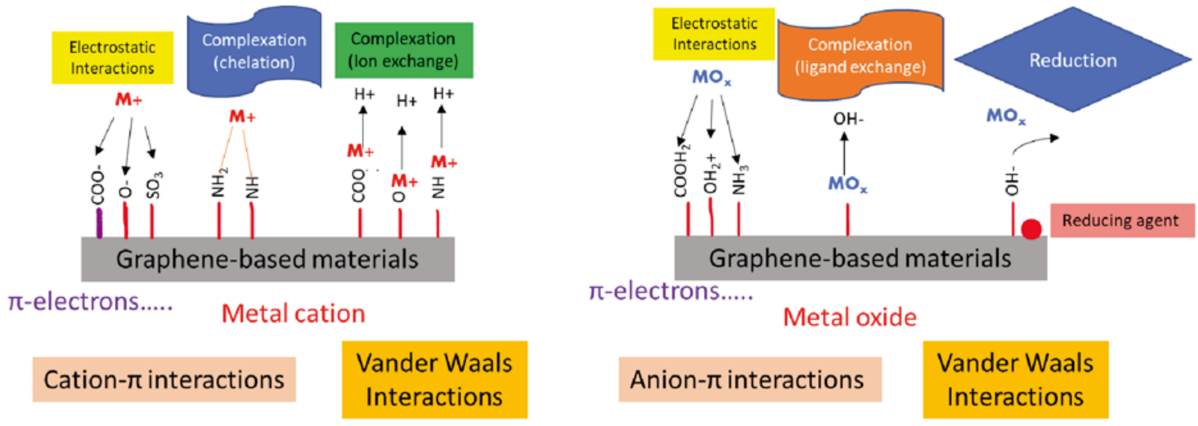

Figure 5. Possible interacting forces for pollutants removal by GO[63]

graphene-based adsorbents. Because heavy metal cations typically adsorb in slightly acidic or neutral solutions, the process of stripping the cations is then carried out in the opposite direction using powerful acids like HCl and HNO_3 [128]. After regeneration, the adsorbent made of graphene can be used again and again without suffering a substantial loss in adsorption efficiency, though a small decline in adsorption performance is observed after every cycle. The effectiveness of the adsorbent made of graphene in removing water after each use can have a significant impact on how well they recycle. Enhanced water dispersion of powder materials built on graphene increases the attraction of adsorbents for heavy metal ions but may reduce the effectiveness of their recovery due to adsorbent material loss during recycling steps like filtering and centrifugation. The substantial number of widened and interlinked pores found in 3D-structured adsorbents allow them to sustain a wide adsorption surface area, while the macroscopic assembly protects the adsorbent's structure during subsequent adsorption-regeneration processes and minimizes material loss[129, 130].

CONCLUSIONS AND FUTURE PERSPECTIVES

This study provided an overview of the various GO nanostructured materials' properties, processing, and structural characterization for the breakdown of organic contaminant in effluent water. To modify graphene planes, materials such as nanoparticles, polymers, biomaterials, and functional groups with oxygen and nitrogen have been used. These materials provide a lot of binding sites or a lot of adsorbent surface area, which improves the performance of pollutant removal. The presence of various functional groups on the GO molecules, they can remove multiple kinds of pollutants at once. Electrostatic interactions, complexation, interactions, hydrogen bonds, hydrophobic interactions, and van der Waals interactions have all been proposed as potential interaction processes for the adsorptive removal of pollutants. Adsorption is considered the best technique to remove both dyes and heavy metals from wastewater system. However, electrostatic interactions are more prominent in heavy metal removal. Graphene oxide-based nanocomposites hold significant promise for the future of wastewater treatment because of their unique properties and versatile applications.

Graphene oxide nanocomposites have a high surface area and abundant functional groups, making them highly effective in adsorbing various contaminants from wastewater, such as heavy metals, organic pollutants, and dyes. Future research should likely focus on optimizing these materials to achieve even greater adsorption capacities, making them more efficient in treating a wide range of pollutants. Researchers are exploring methods to modify GO nanocomposites to exhibit selective adsorption for specific contaminants. Moreover, this selectivity can reduce the need for multiple treatment steps, thus making wastewater treatment more cost-effective and efficient. As the understanding of GO and nanocomposites improves, scientists will develop tailored nanocomposites designed for specific wastewater treatment needs. By adjusting the composition and structure of nanocomposites, they can target particular pollutants and optimize the treatment process.

Most importantly, graphene oxide-based nanocomposites can be regenerated and reused for multiple treatment cycles, reducing waste and operational costs. Future work should seek to improve the regeneration techniques and assess the long-term performance and stability of these materials. With the development of new technologies, graphene oxide nanocomposites can be integrated with other advanced technologies such as membrane filtration, photocatalysis, and electrochemical processes. Combining these technologies with graphene oxide-based nanocomposites can enhance the overall efficiency of wastewater treatment processes. For large scale application, the cost of GO based nanocomposites should be reduced over time as production methods are refined and economies of scale are realized. This reduction in cost can make these materials accessible for larger range of wastewater treatment applications. In conclusion, GO nanocomposites hold tremendous potential for revolutionizing wastewater treatment by offering a sustainable, efficient, and versatile solution for water purification. As research and development efforts continue, these materials are likely to play a pivotal role in addressing the global challenge of wastewater management.

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