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Review Article

Graphene Oxide-Based Nanomaterials: The Preparation, Application, and Factors that Affect the Adsorption Capacity on Drinking Water Treatment-Review

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Abstract

The presence of contaminants in the water is a problem that worries humanity since the consumption of water with inadequate concentrations of contaminants can cause serious problems to human health, and different technologies have been used to reduce or eliminate those contaminants in drinking water. Adsorption has been highlighted because of its low cost and simplicity removal process and, graphene oxide has been reported as excellent adsorbent because it has functional groups that offer stability, antifouling, and hydrophilicity, important to contaminants removal from water. The review of literature related to graphene oxide, water contaminants, and their impact on human health was the basis for the execution of the present work. A thorough analysis of each selected article and critical analysis were made to report the ability to remove contaminants and factors that can contribute positively or negatively to the contaminants removal process. A total of 204 articles were selected, mostly published in the last 5 years, because advances in the use of graphene oxide as adsorbent are reported annually. This article will help to understand better that graphene oxide alone or combined can treat different contaminants from drinking water.

Keywords: Graphene oxide; Adsorption; Organic, Inorganic; Biological; Radiological contaminants

1. Introduction

Water is a crucial element of human survival [1, 2] and development [3]. Clean water is a finite natural resource, and water consumption is soaring due to the rapid growth of the population [4, 5], industrialization [6-14] and critical disposition of water pollution [15]. Because of the problems illustrated in the current paragraph, the majority of issues that humankind is facing in this area are interlinked to the water quality or quantity concerns and, owing to these water problems, the severe impact on the human health includes lack of better sanitation, exposure to the pathogens via the recreation or food chain, etc. [16].

In recent years, innovative solutions have been devised in attempts to solve these issues for water environmental remediation [17]. For example, graphene-based nanomaterials [18] have exhibited many exciting properties. These include adsorption of metal and organic dyes, antimicrobial capability, and photocatalytic degradation of organic molecules [19]. Because of its unique structure (two-dimensional material) [20-30], graphene oxide (GO) have been intensively investigated [31], for drinking water treatment process.

GO can be obtained from the oxidation of graphite [32-34] and its exfoliation [35]. GO is conventionally prepared by chemical oxidation of graphite using Brodie, Staudenmaier, Hummers methods [2,36-41], or some variations of these methods and subsequent exfoliation of graphite oxide using various reduction techniques like thermal, microwave, laser, etc. [42]. Graphene oxide can be combined with other nanomaterials such as CuO [43], Fe−Mg (hydr) Oxide [44], FeOx [45], MoS_2 [46], Ag [47], TiO_2 [48], Fe_3O_4 [49], ZnO [50], to form nanohybrids material, to remove contaminants from water efficiently. The present work will focus on collecting relevant information from studies already done related to the most used graphene oxide preparation methods, its application, and factors that affect the adsorption capacity on drinking water treatment. The work will be useful to the researchers associated with water treatment using graphene oxide because they may have relevant information in this review article.

1.1. General understanding of contaminants in drinking water

Several are the causes (intrinsic and extrinsic) that contribute to the presence of contaminants in water. Ingesting unrecommended contaminant concentrations such as biological, organic, inorganic, and radiological can create collateral damage to living organisms such as plants, humans, animals, etc. Inorganic contaminants such as Fluoride, arsenic [51], lead, copper, mercury, chromium, cadmium [52, 53] may have geological or anthropogenic, natural deposits and industrial practices [54] as sources and, can cause health consequences such as cancer, human health, aesthetic [16] and skeletal deformity [51]. Organic contaminants such as pharmaceuticals [55], pesticides, synthetic and natural hormones, industrial compounds, and personal care products, among others [56], usually the sources are agricultural, public hygienic, industrial and hospital, can cause cancer, hormonal disruptions and nervous system disorder [54] to the human body. Biological contaminants such as pathogenic organisms (bacteria, protozoan, and viruses) [53] are often caused by human and animal fecal waste [57], wastewater from sewage [54]. The contaminants' presence in drinking water can cause dysentery, cholera, and gastroenteritis can damage the nervous system (neurotoxins) and skin [54]. Radiological contaminants such as Uranium [58], alpha particles, beta particles, and photon emitters, radium 226 and radium 228 (combined) [54] can come from natural sources [58], some industrial waste [54] and can cause cancer [58], skin and nervous system toxicity [53]. In particular, graphene oxide (GO) nanosheet (oxygenated graphene sheets bearing carboxyl, hydroxyl, and epoxide functional groups) offer an extraordinary potential for making functional nanocomposite materials with high chemical stability, strong hydrophilicity, and excellent antifouling properties, all of which are promisingly exploited in water treatment processes. Most water-related applications of Graphene oxide have focused on its adsorptive [19] mechanism to remove different contaminants.

2. Methodology

The objetive of the review is to understand better the synthesis of graphene oxide, its evolution, and the different possible applications in removing biological, organic, inorganic, and radiological contaminants in the water. A total of 204 articles were mostly obtained through the Web of Science and analyzed, discussed, and compared according to the type of contaminants to be removed from the water and consider different factors that may improve water pollutants removal. A critical analysis of some literature was also made based on the experience and detailed explanation reported by other authors in different literature works. To better report up-to-date information, of the 204 articles selected, the overwhelming majority were published between 2015-2020.

3. Graphene Oxide Evolution and Preparation

Several methods are used to produce graphene oxide, which provides distinctly different materials with a significant difference in the relative abundance of various functional oxygen-containing groups. The most common of them are Hummers and Brodie's methods [59-61], Staudenmaier method [62-64], Hofmann method [65, 66], and other modified methods such as modified Staudenmaier's method [67], modified Hofmann method [68], modified Hummer [69-74] and improved Hummer [75-77]. These standard methods have evolved since their discovery to the current date (see Figure. 1); they differ from each other because of the involved chemicals, oxidation time, and environmental safety; for example, C. Schafhaeutl, Brodie, Staudenmaier, and Hofmann's methods use strong chemicals, produce toxic gases and long oxidation time, compared to Hummers and Improved hummers methods. The choice of the procedure to be used depends on each methods advantages and disadvantages (see Table 1) and the purpose of the produced GO.

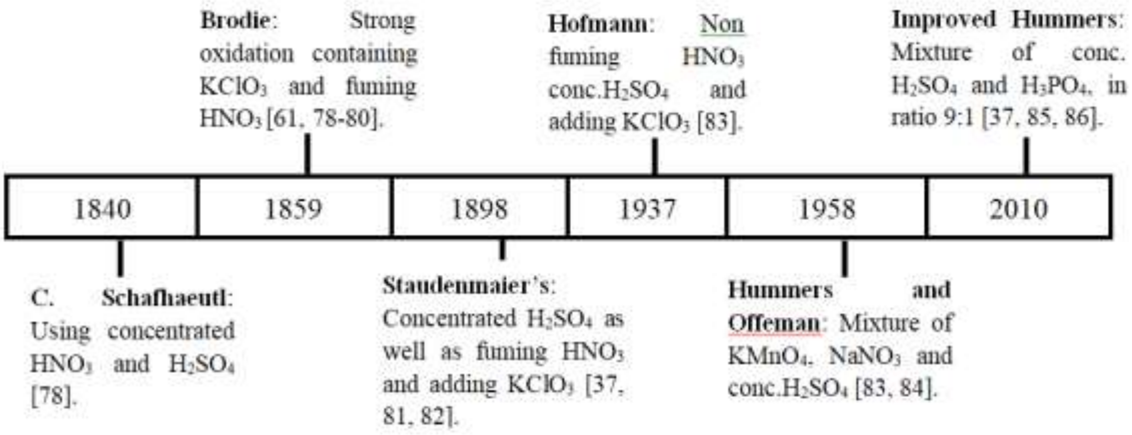

Figure 1: Evolution and difference between methods used to prepare GO using chemical oxidation.

Since the works of literature, [87-91] and [92] shows the synthesis procedure of Graphene oxide using Brodie method, Staudenmaier method, Hofmann method, Hummers method, Marcano Improved Hummers method and modified Hummers method respectively. They provide distinctly different materials with a significant difference in the relative abundance of various functional oxygen-containing groups, significantly different properties [59], and different structures obtained. The distribution of the types of oxygen-containing groups available could affect the performance of the GO (or its exfoliated form, graphene oxide) materials in specific applications [83].

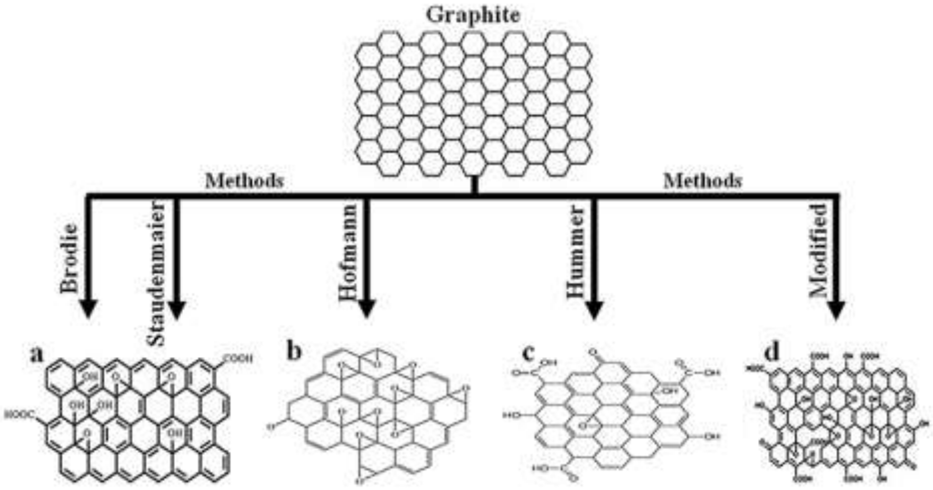

Figure 2: Structure of graphene oxide produced using **(a)** Brodie and Staudenmaier methods, reprinted with the permission of [93]; **(b)** Hofmann method, reprinted from the literature [94] with the permission of Spring Nature and Copyrights Clearance Center – license number 4917670969786; **(c)** Hummer method, reprinted from the literature [95] according to MDPI policy and; **(d)** Modified method, reprinted from the literature [96] with permission of Elsevier and Copyrights Clearance Center – license number 4917460128617.

Different structures of Graphene oxide (see Figure 2) produced using different methods is shown a few defects [59] and less oxygen [82] (see Figure 2a), more oxygen compared to Figure 2a (see Figure. 2b) and a higher proportion of C=O bond type (see Figures 2c and 2d). The O−C=O bond type occurrence indicates a significant presence of the carboxyl group [83].

Methods	**Advantages**	**Disadvantages**
C. Schafhaeutl	The earliest method [78].	Not clear about the type of graphitic oxidation [78].
Brodie	Less aggressive [97].	Very long reaction time-up to several days; Have certain risk of self-ignition or explosion [98]; Lower sorption capacity and lower activity [59].
Staudenmaier	Highly oxidized Graphene oxide in a single reaction vessel [99], compared to Brodie method.	Large particles and lower interlayer spacing [100]; Hazard and time consumer [101].
Hofmann	Avoid the usage of corrosive fuming HNO_3 [102]; High degree of oxidation, small particles, compared to Staudenmaier method [100].	Risk of explosion [65, 103, 104].
Hummer	Few hours to complete the reaction; Avoid the evolution of explosive ClO_2 [105]; Eliminate the formation of acid fog [106]; Non-toxicity [107, 108], compared to above methods.	Toxic gases are produced [105, 109, 110]; The residual Na^+ and NO_3^- ions are difficult to be removed from wastewater formed [106]; Allows the diffusion of oxidizing agents into the interlayer spaces of graphite [111].
Improved Hummer [37].	Eliminate the generation of toxic gases, avoiding the $NaNO_3$ [106, 112-114]; Large amount of oxidized graphite is provided [115-117].	Separation and purification processes are tedious processes [115].
Modified Hummer	Improved level of oxidation and, therefore, product performance [115].	Production of NOx gases [102, 115].
Improved method [118].	Less acid used; Reduce the cost of waste-acid treatment as well as lowering the impact on the environment.	The temperature must be well controlled to reduce the safety risk.
Simplified Hummer [119].	Oxidized graphene without the use of a strong oxidation agent.	-

Table 1: Advantages and disadvantages of the Graphene oxide preparation methods, including some hummer's modified methods.

4. Strategies for Graphene Oxide Application

Many techniques have been utilized for water purifications, in which the adsorption is known as a simple, effective, and economical method to achieve high-performance water purification due to its high ability to remove various soluble and insoluble organic and inorganic contaminants [120]. Graphene-based nanocomposites and its derivatives [52] are developed as the impressive adsorbent materials owing to its outstanding features [121]. Graphene oxide (GO) as the most studied graphene-derivatives, is a precursor material in graphene preparation and has a large number of carboxyl, hydroxyl [122-124], carbonyl, and epoxy carboxyl groups on its surface [125-127], which are responsible for binding both organic and inorganic species [128]. Graphene oxide is hugely hydrophilic and has oxygen functional groups on the structure [8, 129-137], making it ideal to be used in aqueous solution [125]. Water flow is afforded due to the spacing between nanosheets, which is typical of the order of 0.3-0.7 nm [138]. It is ideal for water permeation while blocking the transport of larger molecules [139]. Figure 3a shows the adsorption mechanism that graphene oxide has. It is possible to see that only water can pass from the graphene oxide layer; the large and small particles are blocked and not allowed to pass the GO layer.

Many studies are made to prove the significant advantages that graphene oxide has in removing the contaminants from water. Graphene oxide, by itself, is an attractive adsorbent and becomes increasingly interesting in the removal of contaminants when combined with other nanomaterials, forming nanohybrids. To better understand the importance of graphene oxide in the contaminants removal process, table 2 shows different technologies and performance of graphene oxide to remove contaminants from the water.

4.1 Strategies for using graphene oxide to remove bacterias from water

Pathogens are microscopic, single-celled organisms that thrive in diverse environments, including water. To remove the harmful ones, LIU et al [140] studied the antibacterial activity of different graphene-based materials. They found that the produced graphene oxide had higher antibacterial activities than other studied materials (graphite oxide, graphite, and reduced graphene oxide), under similar concentration and incubation conditions. According to them, the reason for GO's excellent bacteria adsorption is related to the size [141] of diameter (small diameter has higher antibacterial activity than larger diameter). Another reason is related to stable dispersions with small nanosheets (because of carboxyl, hydroxyl, and epoxy groups introduced on graphene oxide sheets), thus offers more opportunities to interact with cells for cell deposition. They showed that the incubation time and nanomaterials concentration to remove E. coli from water have a relationship. The loss viability increases from 49.1 ± 6.0% after 1h incubation to 69.3 ± 6.1% after 2h, and further increases to 81.5 ± 3.9% after 3h and 89.7 ± 3.1% after 4h. The loss of E.coli viability jumps from 10.5 ± 6.6% at the graphene oxide concentration of 5 µg/mL to 91.6 ± 3.2% at 80 µg/mL. The majority of E. coli was killed after incubation with graphene oxide at 80 µg/mL concentration.

Ming et al [142] studied the adhesion of bacteria to a graphene oxide film. They tested the interaction between graphene oxide film with several bacteria, such as Escherichia coli (E. coli), Staphylococcus aureus (S. aureus), Shewanella-MR 1 (Shewanella), and Bacillus. They studied the ability to graphene oxide to get electrons to adhere to bacteria (see Figure 3b). As an electron acceptor, graphene oxide was easier to get electrons from Shewanella than that of the other bacteria studied; it was why Shewanella's adherence to the GO film was low (below 3%). Moreover, the strategy of impressed current provided new insight into the increase in the adherence of bacteria. In this study, they also found that exerting a positive current would draw the electrons from graphene oxide and enhance graphene oxide ability to obtain electrons from bacteria, resulting in effective adsorption of bacteria on graphene oxide films. The ease that graphene oxide has to adhere to bacteria comes from the nature of graphene oxide to get electrons from bacteria. They also reported the dependency between the mass of graphene oxide and adherence effects toward different bacterias. Was possible to understand that when the graphene oxide film mass was in the range of 0.398mg to 3.184mg, the adherence percentages for E.coli changed from 20% to more than 100%, while they changed only from 18.4% to 31.4% for S.aureus, from 12.0% to 52.3% for Bacillus, and from 0.8% to 3.1% for ShewanellaMR-1.

Akhavan and Ghaderi, [143] studied the toxicity against bacteria of graphene oxide nanowalls achieved by electrophoretic deposition of Mg^{2+} graphene oxide nanosheets. They reported that the bacterias were effectively damaged by direct contact with the nanowalls, very sharp edges of the, resulting in inactivation of the bacteria by the nanowalls; The cell membrane of S. aureus bacteria was more damaged, compared to the cell membrane of E. coli. Assigned the more resistance of the E. coli bacteria against the direct contact interaction with the nanowalls as compared to the S. aureus bacteria to the existence of an outer membrane in the structure of Gram-negative E.coli bacteria and the lack of such an outer membrane in the form of Gram-positive S. aureus bacteria.

Rajapaksha et al [43] studied a hybrid GO-CuO to remove waterborne pathogenic bacteria (E. coli and S. Typhimurium bacteria). The resulting GO-CuO nanocomposite was found to be an extremely effective antibacterial nanomaterial, significantly inhibiting the growth of Escherichia coli and S. Typhimurium bacteria. The GO-CuONP nanocomposite revealed a moderate concentration-dependent antibacterial activity, with the inhibition increasing as a function of substrate concentration. The observed GO-CuONP could dramatically inhibit the growth of E. coli and S. Typhimurium. The study shows that the ideal concentration of GO-CuONP for S. Typhimurium removal was 2mg/ml and 3mg/ml to remove E.coli; the removal capacity was ~98% and ~88%, respectively. The antibacterial activity of the GO−CuONP nanocomposite could arise from the combined effect of both the native graphene oxide and the surface deposited CuONP. However, it appears that the derived activity is mostly affected by the inherent antibacterial of the CuONPs; this means that the GO primarily acts as a scaffold structure. Were widely demonstrated that the results obtained are unsurprising because of the antibacterial activities of both CuO [144-147] and GO [148], are known.

Song et al [149] studied the antibacterial properties of GO-Ag nanocomposites. Even requiring more investment, the combination exhibited great antibacterial activity, and the bactericidal process was controlled within 25 minutes. They found that the antibacterial behavior of GO-Ag against both bacterial strains (E. coli and S. aureus) was not only a dose-dependent process but also a time-dependent contact one. At 25 minutes, the antibacterial rates for E. coli reached 89.72%, 97.83%, 99.99%, and 99.99%, while the antibacterial rates for S. aureus were 70.32%, 95.53%, 95.70%, and 97.65%, when the concentrations of GO-Ag were 40, 120, 200 and 280 mg/L, respectively. GO-Ag was much more destructive to the cell membrane of E. coli than that of S. aureus, which could also provide evidence for the result mentioned above that antibacterial activity of GO-Ag against E. coli was more effective. Cell membranes were then first to be affected when the cells were exposed to harmful material. Negatively charged lipids quickly took up positively charged silver ions on cell membranes because of electrostatic attraction.

Li et al [150] studied the multifunctional combination between Lysozyme (Lys), Tannic acid (TA) and GO, to remove bacterias from water. The results displayed high efficiency on the killing bacteria, both Gram-negative bacteria (E. coli) and Gram-positive bacteria (S. aureus), and enhanced osteogenic differentiation. The results obtained can be justified by the natural characteristics from used materials; for example, graphene oxide kills bacteria through multiple ways: cutting the bacteria membrane by its sharp edge, inducing oxidative stress and, TA kills the bacteria through several mechanisms too, such as interacting with biomolecules and metal ions within bacteria, increasing the cell membrane permeability, destabilizing the cytoplasmic membrane, and changing protein to lipid ratios in the membrane. The incorporated GO, Lys, and TA play multimode in killing bacteria (see Figure 3c) because of their respective and different antibacterial mechanism. Under such multiple and different antibacterial mechanisms, if even one component was out of action, the antibacterial effect could be offset by another element, leading to high efficiency in killing bacteria.

4.1.1 Effect of pH to remove bacterias from water:

In addition to the contact time, size, and the concentration of the material in use, it was reported by Song et al [149] that the GO-Ag showed better antibacterial activity against both E.coli and S.aureus bacteria under the acidic condition in comparison to the disinfection of the same water at higher pH value. For E.coli, the antibacterial rates were 81.68%, 75.5% and 70.5%, and for S.aureus, the antibacterial rates were 58.23%, 45.96% and 40.6%, when the pH values were 5.5, 7 and 8.5, respectively. Different concentrations of silver ions released from AgNPs at varying pH values can explain the obtained results.

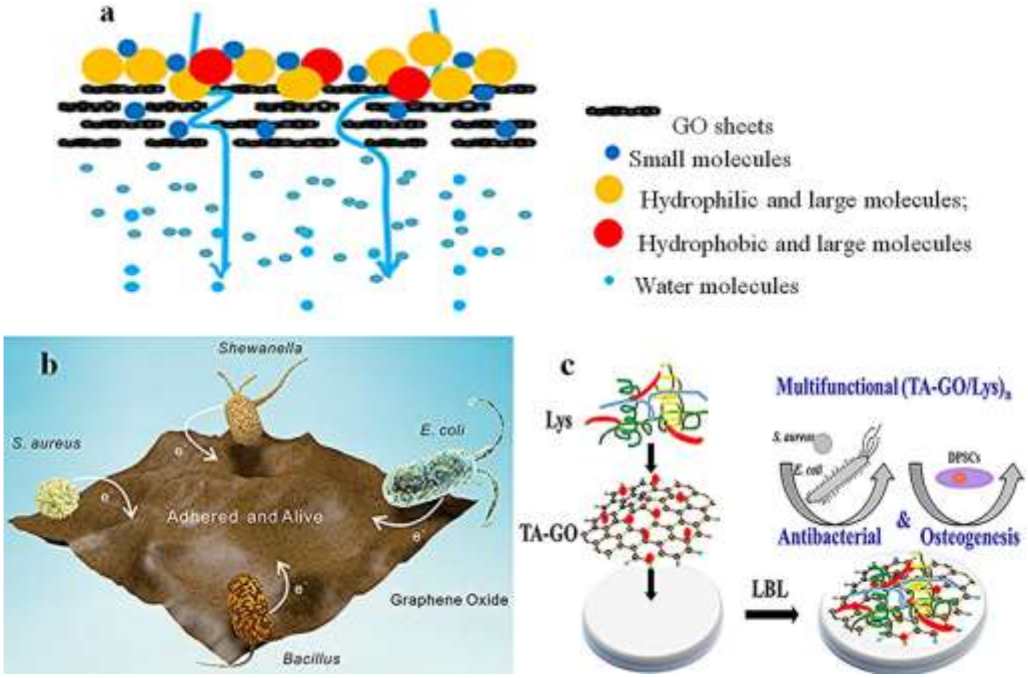

Figure 3: **(a)** Mechanisms of GO adsorption, reprinted and modified from the literature [31] with permission of Elsevier and Copyrights Clearance Center-license number 4917471105182; **(b)** Schematic diagram of bacteria adhered on GO, reprinted with permission from literature [142], Copyright 2020, American Chemical Society, and; **(c)** schematic diagram of multifunctional (TA-GO/ Lys)n to remove bacterias, reprinted with permission from literature [150] Copyright 2020, American Chemical Society.

4.2 Strategies for using GO to remove different inorganic contaminants from water

4.2.1 Heavy metals removal from water: Among the inorganic contaminants, heavy metals [151] and metalloids are the most harmful because they have high toxicity at low concentrations [152, 153]. Metal ions can easily interact with graphene derivatives because of the π-systems and carboxyl and hydroxyl groups on the edges and surfaces [154-156]. Graphene oxide has been studied to remove different types of inorganic contaminants from the water. Ain et al [74] have reported that magnetic graphene oxide can remove heavy metals (Pb^{+2}, Cr^{+3}, Cu^{+2}, Zn^{+2}, and Ni^{+2} ions) from water. The carbon material ability to adsorb metal cations from the solution depends on a range of parameters, including the mixture's pH and ionic strength [157]. The adsorption capacity of magnetic graphene oxide (MGO) was found to be 200 mg/g for Pb^{+2}, 24.330 mg/g for Cr^{+3}, 62.893 mg/g for Cu^{+2}, 63.694 mg/g for Zn^{+2} and 51.020 mg/g for Ni^{+2}. The MGO increases by increased pH. High pH, the functional groups are deprotonated, and the surface of MGO becomes more negatively charged. More metal ions are attracted and attached to the surface of MGO due to the strong electrostatic [158] attraction, and the removal of efficiency is increased.

4.2.2 Arsenic removal from water: Arsenic is one of the highly toxic and carcinogenic chemical elements, and its contamination in natural waters has become a global problem [159]. Among the four different oxidation states of arsenic, As^{3+} is ten times more toxic than As^{5+} and seventy times more toxic than methylated arsenic compounds [160]. To remove those toxins from water, Wu et al [161] synthesized a magnetic Fe@Cu&GO composite by successive growth of Fe_3O_4 and CuO on the surface of GO sheets to remove arsenic (III/ V) from water and, they saw that graphene oxide increased the adsorption rate of As(V) and As(III) compared with Fe@Cu adsorption rate. They also compared the adsorbents results obtained from other research and concluded that the combination they made (Fe@Cu&GO) presents better As(III/ V) removal from water. The maximum amounts of adsorbed As(III) and As(V) were 70.36 and 62.60mg/g, respectively. Pourbeyram et al [162] found high amounts of adsorbed As^{3+} and As^{5+} in the combination of graphene oxide and zirconium (GO-Zr) nano-composite. The maximum adsorption capacity found in the study was 212.33mg/g and 232.52mg/g for As^{3+} and As^{5+}, respectively.

β-FeOOH incorporated carboxylic graphene oxide nano-composite β-FeOOH@GO-COOH nano-composite has removed 100% arsenic ions from water. The composite has shown tremendous adsorption efficiency even after 20 successive adsorption-desorption cycles and adsorbed >80% of both As(III) and As(V). The maximum adsorption capacity for β-FeOOH@GO-COOH was found to be 77.5 and 45.7 mg/g for As(III) and As(V) ions, respectively. Figure 4a shows the bonding between β-FeOOH@GO-COOH, As(III), As(V), and the stretching vibrations of As–O group in the As–O–Fe linkage at the surface of the β-FeOOH@GO-COOH composite. The adsorption behavior depends not only on the charge properties of the adsorbent surface but also on the specific interactions between functional groups on the adsorbent surface and the adsorbed species [163].

4.2.3 Mercury removal from water: Due to its volatility, persistence, and bioaccumulation, mercury (Hg) has been considered as one of the most toxic metals [164], which can affect the health of human beings [165]. To remove mercury from polluted water, Bao et al [166] reported in their research the higher Hg^{2+} adsorption capacity of Thiol-functionalized magnetite/ graphene oxide (MGO). Its ability reached 289.9 mg/g in the solution with an initial Hg^{2+} concentration of 100mg/l. The adsorption capacity could be attributed to the affinity of Hg^{2+} by magnetite nanocrystals and thiol groups. Henriques et al [167] observed that GO macrostructures are a beneficial material on the sorption of Hg(II) from water solution. The different chemical surface modifications performed showed that the combination of oxygen and nitrogen functional groups (3DGON) increases by 96% the removal efficiency of Hg (II).

Kabiri et al [168] studied a functionalized graphene-based composite with a unique 3D architecture composed of graphene nanosheets decorated with αFeOOH nanoparticles and porous diatom silica microparticles and, the results showed high-efficiency adsorption of Hg ions in water. The adsorption performance for Hg removal was achieved with an adsorption capacity of >800 mg/g (at 400 mg/l Hg^{2+}). Rathore and Biswas [169] showed that GO@SnS_2 exhibits highly selective and efficient removal of Hg(II) from the water with a capacity of 342.02 ± 8.02mg/g. The highly accessible Hg(II) binding sites, S^{2-} from SnS_2 and COO– on the GO surface, play a synergistic role in the effective Hg(II) capture. It can sequester 99% Hg(II) selectively even in the presence of Na(I), K(I), Cs(I), Rb(I), Mg(II), Co(II), Cu(II), Ni(II), Zn(II), Pb(II), Cd(II), Mn(II), Fe(III) and As(III) with an extremely high separation factor.

4.2.4 Fluoride removal from water: Fluorine, one of the 14 necessary elements to the human body and has a positive effect on human in a specific concentration (0.7mg/l), but excess fluoride intake (1.5mg/l) [170] is harmful to human [171] and aquatic organisms [172]. To Remove fluoride ion from polluted water using GO, Chakra et al [173] reported in their study the combination between GO and zinc oxide nanocomposite. The adsorption capacity of GO-ZnO nanocomposites for fluoride was 16.608 mg/g. The increase in the removal of fluoride point outs that the fluoride adsorption perhaps occurs due to the diffusion of intriguing position inside the pores on the adsorbent surface. The fluoride ions removal from the water rises with an increase in agitation time to a small degree. Prabhu et al [174] reported an alternative adsorbent for fluoride removal, assembling nano-sized hydroxyapatite onto graphene oxide (GO-nHAp). 44.068mg/g was said to be the highest adsorption. Liu et al [175] reported the maximum adsorption capacity of 17.67mg/g in the study about defluoridation by rice spike-like akaganeite anchored graphene oxide. Shang et al [176] enhanced fluoride uptake by bimetallic hydroxides (Zr-La) anchored in cotton cellulose (CC)/ graphene oxide composites (Zr-La-CC/GO). They reported that fluoride uptake was significantly increased from 45.1mg/g to 81.3mg/g with an increased pH from 2.5 to 3.0. Zr-La-CC/GO nanocomposites exhibited higher fluoride adsorption at lower pH conditions. It was reasoned that it could selectively adsorb the fluoride ions onto the positively charged Zr and La hydroxides and formed Zr-F and La-F complexes in the inner-sphere composites (see Figure. 4b). They protonated the Zr/La species in an acidic environment, which favors F^- or HF adsorption based on electrostatic attraction. At the equilibrium stage, the fluoride uptake capacity onto Zr-La-CC/ GO nanocomposites was about 38.8mg/g. Wang et al [177] studied the synthesis of $(ZrO_2\text{-}Al_2O_3)$/GO nanocomposite for high defluoridation. The results show a maximum fluoride adsorption capacity of 62.2mg/g and adsorption ability of 13.80mg/g when the F^- equilibrium concentration is 1mg/l. Figure 4c indicates that all three components (ZrO_2, Al_2O_3, and GO), prepared by the sonochemical method and incorporated together, can significantly improve adsorption capacity.

4.2.5 Copper removal from water: Cu^{2+} ion is an essential ion in biological systems, such as various redox processes and enzyme functions. Deficient or excessive Cu^{2+} ion levels are usually associated with some severe disorders [178]. Copper excess may severely affect the ecological cycle and, subsequently, human health [179], and the recommended concentration in drinking water has to be below 2mg/l [180]. To maintain potable water with recommended copper concentrations, Huang et al [181] studied a molecular beacon and GO-based fluorescent biosensor for Cu^{2+} detection; Zhang et al [182] studied the synthesis of nitrogen-functionalized graphene oxide for copper adsorbent; Wang et al [183], learned a graphene quantum dots as a fluorescent sensing platform for highly Cu^{2+} detection and, Shao et al [184], studied a nanoscale zero-valent iron decorated on bentonite/ graphene oxide for removal of copper ions. They found ~50nM as detection limit, 26.7451mg/g as adsorption capacity, 0.226µM as detection limit, and 184.5mg/g as maximum adsorption limit, respectively.

4.2.6 Other important factors to consider to remove inorganic contaminants from water

4.2.6.1 Effect of contact time: The contact time required to achieve more excellent performance to remove heavy metals from water depends on the combination of materials or the type of material used. For example, Ain et al [74] reported that the best results of removing heavy metals (Pb^{+2}, Cr^{+3}, Cu^{+2}, Zn^{+2}, and Ni^{+2} ions) from water, was verified from 25 to 35 minutes. The study reported by Pourbeyram et al [162] shows that the adsorption of As(V) on the nanocomposite increased rapidly during the first 5 minutes and gradually increased and finally reached equilibrium after 10 min. In the same condition, the removal of As(III) took some more time than As(V). The study made by Rathore and Biswas [169] reported results close to 100% of Hg(III) removal, although the long contact time required (close to 1500 minutes) to achieve the maximum reduction of Hg(III). The reference [173] illustrates fluoride removal in the range of 0-240 minutes. The fluoride percentage values were escalating linearly up to 40 minutes; afterward, it remains nearly invariable, signifying the accomplishment of adsorption equilibrium; suddenly, at 200minutes, it showed an increase in value up to 240 minutes. Therefore 240 minutes showed the highest defluoridation (83%). The study made by Prabhu et al [174] reports an increased defluoridation capacity in the contact time from 5 to 25 minutes, resulted in an increase in the adsorption capacity from 78% to 96%, which indicates that the number of active sites gradually occupied. The sorbent's defluoridation capacity was almost saturated at 25 minutes and reached a maximum of 96%.

4.2.6.2 Effect of pH: pH is the most crucial factor in the liquid-solid adsorption procedure [162]. To get satisfactory results in removing pollutants, it is essential to know the ideal pH to be used. It's important to realize that the same or different materials may require different pH to remove various water contaminants. The study from reference [161] demonstrates the adsorption capacity of As(III) and As(V) over a pH range of 3 to 10 and reveals that the use of different pH can efficiently remove As(III) and As(V). For example, to remove the maximum of As(V) was necessarily verified an acid pH (pH=3), and the maximal removal of As(III) was with pH in the range of 8 to 10, which was explained by the possibility that more OH^- competed with arsenate anions for active sites at higher pH conditions. The effect of fluoride adsorption shown by Prabhu et al [174] indicates that the maximum fluoride uptake between the pH ranges 3–7 reached saturation. The pillar-like support of graphene oxide materials would make the benefit of the growth of hydroxyapatite materials using a large number of functional groups like -OH, -COOH through hydrogen bonding and van der Waals forces of attraction which would attract more fluoride ions in acidic condition by electrostatic attraction (96%). The adsorption capacity reported by Liu et al [175] towards fluoride showed a similar uptake rate when pH values ranged from 2.1 to 10.4, with the maximum adsorption rate around 80%. The adsorption of fluoride results in an apparent drop from pH 11.1 to 12.7, even dropping from 45.8% to17.6%. In the removal of Cu(II) and Cr(VI), Kumar et al [179] reported that the best results of adsorption capacity were ~200mg/g and ~340mg/g, with pH=5 and pH=3, respectively. According to reference [184], the pH of the solution was among the most critical factors that affected the removal efficiency of Cu(II) for the surface speciation of the adsorbent in contact with the solution and speciation of Cu(II) in solution. The removal efficiency of Cu(II) increased with the change in pH from 2 to 6. They also found that Cu(II) absorption depends mostly on pH, with higher sorption of cations at high pH and higher sorption of anions at low pH. The removal efficiency of Cu(II) was very low; only 3% to 10% of Cu(II) had been removed when the pH was 2. One of the main factors was that fewer surface active sites of adsorbents could be accessible to Cu(II) because at low pH was possible to see the competition between Cu(II) and (H^+). In acidic environments, iron particles can be corroded by (H^+), resulting in fewer iron particles reacting with Cu(II). When the pH value of the solution was 5, the removal efficiency of Cu(II) reached a relatively high result (86%), and the maximum adsorption was 93% at pH=6.

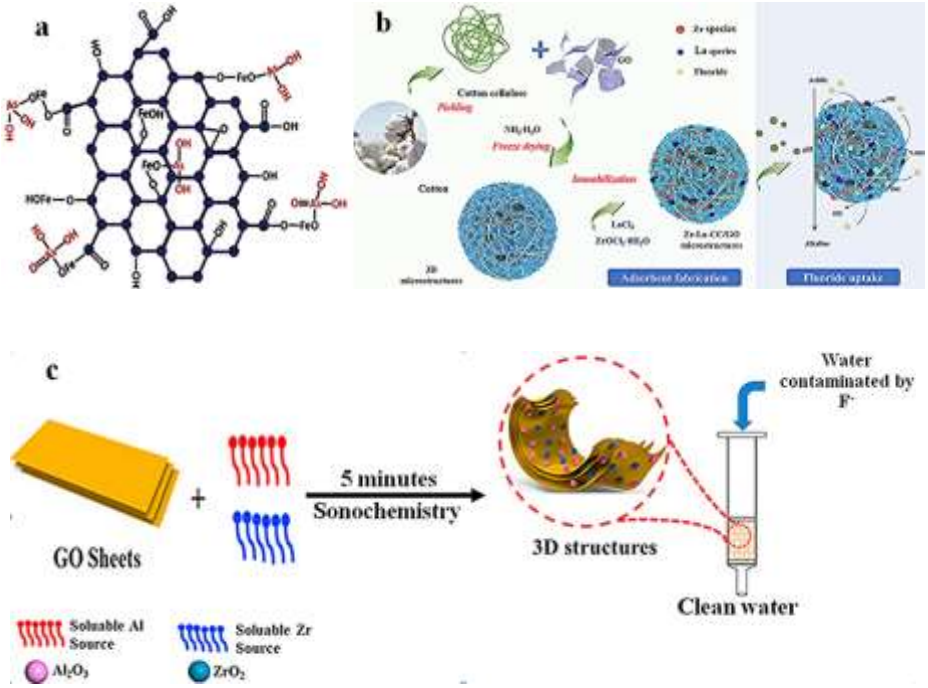

Figure 4: **(a)** bonding between β-FeOOH@GO-COOH, As(III) and As(V), reprinted from reference [163] with the permission of Elsevier and Copyrights Clearance Center – license number 4918011449709; **(b)** Zr-La-CC/GO nanocomposites fabrication and adsorption schemes, reprinted from reference [176] with permission of Elsevier and Copyrights Clearance Center – license number 4917470394440; **(c)** (ZrO_2-Al_2O_3)/GO fabrication and its performance, reprinted and modified from reference [177] with permission of Elsevier and Copyrights Clearance Center – license number 4917461236318.

4.2.6.3 Effect of concentration: Henriques et al [167] showed a direct proportionality between increasing the sorbent concentration and the efficient removal of Hg(II). The results show that higher percentages of Hg(II) removal are obtained with increased sorbent concentration, from 33% to 96% for doses 1 to 20 mg/l, respectively. It was evident that the increase of sorbent mass leads to a larger number of available coordination sites improving the removal efficiency of the sorbent. Whereas, the study reported by Shao et al [184] presents an indirect proportionality with the sorbent and removal efficiency concentration. The removal efficiency of Cu(II) shows that the initial concentration of Cu(II) greatly influenced the removal efficiency. When the initial Cu (II) concentration was 75 mg/l, the removal efficiency was the best, which reached 93%, and the adsorbent decreased with the increase of the initial concentration; this was due to the limited adsorption sites of the iron particles, and before the high concentrations of Cu(II) was adsorbed completely, the iron particles had reached a state of adsorption saturation.

4.3 Strategies for using graphene oxide to remove different organic contaminants from water

Organic water contaminants (e.g., pharmaceuticals and personal care products) are constantly emerging with the rapid advances in various industry sectors. Many of these emerging contaminants are commonly considered micropollutants and have potential adverse health effects [185], thereby raising severe public concerns about water safety [186]. Geng et al [187] studied humic acid removal using Fe_3O_4/TiO_2-N-GO sonocatalyst with ultrasound assistance. The removal efficiency was 80% for sono-adsorption, and 93% for sono-photocatalysis. The increasing removal efficiency was contributed by cavitation effect caused by ultrasonic waves, and resulted in the formation of more OH and other radical species, allowing more humic acid to be removed. Tan et al [188] demonstrated a highly sensitive and selective fluorescent biosensor based on GO-hydrogel for antibiotic detection. GO hydrogels were prepared by physically mixing GO solution with adenosine. The fast gelation of the graphene oxide dispersion in the presence of adenosine may contribute to the electrostatic interactions and strong hydrogen bonding between the adenosine and the GO nanosheets. They related that adenosine contains more than one nitrogen (N)-containing functionality and can accept protons from the carboxylic acid groups (– COOH) of the GO sheets to participate in acid-base-type electrostatic attraction. Adenosine and aptamer were served together as cross-linkers between the GO sheets, as illustrated in Figure 5a. The aptamer can recognize and bind to their cognate targets with high specificity and affinity; these functional hydrogels were prepared primarily to target detection and adsorbed the aptamer probe on the GO sheets via the strong π–π stacking interactions. Thoroughly immersed in the GO-based functional hydrogel in antibiotic solutions, the aptamer would prefer to capture the antibiotic molecule to form a target-aptamer complex (see Figure. 5b). The recoveries were acceptable and calculated to be in the range of 96–117%.

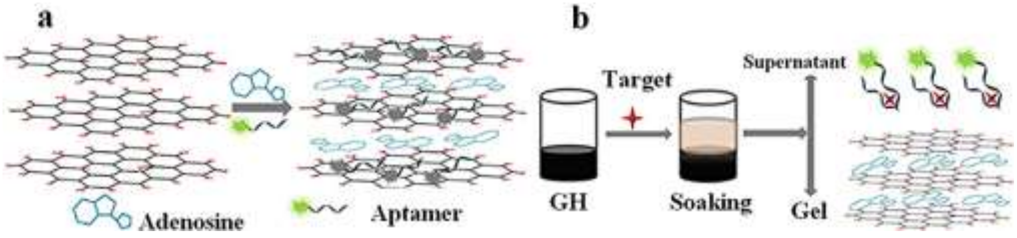

Figure 5: Schematic of the fabrication of **(a)** GO hydrogel; **(b)** the mechanism of selective detection of antibiotics, reprinted and modified from reference [188] with the permission of Elsevier and Copyrights Clearance Center – license number 4917621275241.

4.4 Effect of support membrane to enhance graphene oxide properties in pollutants removal from water

Support membranes always have a place in the field of adsorption [165]; polymer macromolecules, such as polyvinylidene fluoride (PVDF), polyacrylonitrile (PAN), polyethersulfone (PES), polysulfone (PSF) and other materials have been used, owing to their respective advantages [8] such as flux, fouling resistance and life span [35]. The existence of graphene oxide in the membranes induce remarkable properties due to its intrinsic hydrophilicity, compatibility with the polymeric matrices, considerable negative zeta potential, and mechanical and thermal stabilities [10]. Zhao et al [31] reported that the PVDF support membrane by itself could remove dissolved organic carbon (DOC); at 120 minutes, the maximum removal was ~30%. When modified, the PVDF with GO's presence, the DOC maximum removal, was ~70%. They also reported the removal improvement to UV_{254}, and the results were ~58% and ~70% for PVDF and PVDF-GO, respectively. The adsorption amount of membranes increased with the increasing number of GO layers, which might indicate the deposition of GO layers adsorption in the filtration process, which further improved the DOC rejection. It implied that the fouling layers on the modified membranes could improve the DOC rejection through size exclusion and also with increasing filtration time.

Hu and Mi [21] reported a study of graphene oxide membranes' assembly via electrostatic interaction. The hydrolyzed polyacrylonitrile (hPAN) support was first immersed in the poly(allylamine hydrochloride) (PAH) solution to attach positively charged PAH and then in GO solution to deposit negatively charged GO on top of PAH (see Figure 6a), thus completing the assembly of the first GO–PAH bilayer on each side of the hPAN support (see Figure 6b). They used the Characterization techniques to confirm the successful assembly of multiple GO-PAH bilayers, understanding the structure of the GO membrane and quantify their composition and thickness. In low ionic strength solutions, the graphene oxide membrane retained a tight structure and exhibited a high rejection of sucrose (~99%). Wu et al [25] developed a SiO_2–GO nanohybrid/ polysulfone membrane for egg albumin rejections and pure water fluxes. They compared the performance of different inorganic nanoparticles (Psf, SiO_2/Psf, GO/Psf, SiO_2-GO/Psf). They reported that all of the hybrid membranes have a higher water flux than pure PSF membrane, and SiO_2–GO/Psf hybrid membrane exhibited the excellent enhancement. The SiO_2–GO nanohybrid possesses extremely high hydrophilicity due to the synergistic effect of nano-sized SiO_2 and GO. SiO_2–GO nanohybrid demonstrated good dispersion and improved compatibility with the PSF matrix, contributing to the highly maintained egg albumin rejection. They also found that the egg albumin rejection rates of membranes decreased slightly with increasing amounts of SiO_2–GO particles, staying at a relatively high level (>98%).

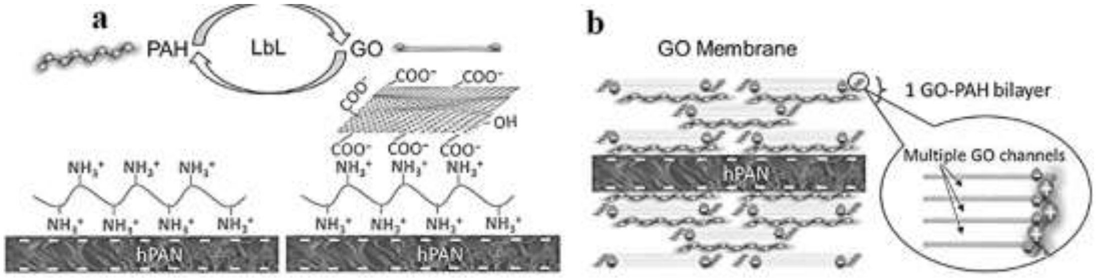

Figure 6: Schematic diagram of **(a)** LbL assembly of a GO membrane by alternately soaking an hPAN support substrate; **(b)** deposit of a prescribed number of graphene oxide–PAH bilayers on both sides of hPAN, reprinted from reference [21] with permission of Elsevier and Copyrights Clearance Center – license number 4917481264067.

Material/ Technology	**Contaminants to be removed**	**Removal capacity: (mg/g) or (%)**	**Ref.**
EDTA-magnetic GO	Pb^{2+} Hg^{2+} Cu^{2+}	96.6% 96.1% 94.3%	[189]
PVK-GO	Pb^{2+}	888mg/g	[190]
GO-Magnetic Chitosan	Zn (II)	71.4 mg/g	[191]
GO-AgNPs-MS	E. Coli S. Aureus	99.8% 99.3%	[192]
SMGO	Uranium - U(VI)	95.2%	[193]
Cu-GOS	E. Coli	>99.9%	[194]
CuO-GO	Nitroaromatics	85%	[195]
GO-Ag	E. Coli	85.6%	[196]
GO-γ-Fe_2O_3	1-naphthol Bisphenol A Atrazine Dibutylphthalate	680mg/g 360mg/g 200mg/g 200mg/g	[197]
TiO_2-GOFe_3O_4	Microcystin-LR	65-100%	[198]
MGO-Sr	Phosphate ions Nitrate ions	238.09mg/g 357.14mg/g	[199]
Ox-GO-Zr	Fluoride	9.70mg/g	[200]
PPY-GONC	Cr (VI)	625mg/g	[201]
GO	TC	323mg/g	[202]
Sp-GO/PLL	E. Coli B. Subtilis	61% 59%	[203]
Fe_3O_4@DTSA-GO	Hg (II)	283.5mg/g	[204]
GO-TiO_2	Carbamazepine Caffeine	99%	[29]

Table 2: Technologies and performance in the removal of pollutants using graphene oxide.

5. Critical Observation

Water is an essential and essential good for living beings. To obtain the potable water (safe for human consumption) still a real challenge for the whole world, especially for the less developed countries. With the discovery of new water treatment technologies using nanomaterials such as graphene oxide, are given different solutions to safely remove contaminants from the water with minimal possible water recontamination. The underdeveloped countries less use advanced technologies to remove pollutants from the water because the solutions proposed by many researchers to purify water are not economically viable. For example, the study showed by Song et al [149] even with fantastic results in removing bacteria, the nanomaterials used require more investment (are expensive); it shows that their study is useful only for developed countries.

Using the same concentration and incubation conditions, LIU et al [140] reported that the graphene oxide showed to be the best nanomaterial to remove bacterias from water; but, the study reveals only the time and concentration-dependent and does there mention about the possible pH dependence, as reported by Ming et al [142]. Akhavan and Ghaderi [143] reported that higher bacterial toxicity shown was related more to sharpening the nanowalls edges. But, Rajapaksha et al [43] contradicting, said that the higher bacterial toxicity demonstrated in a similar study was not related to their sharp edges but related to the mechanism relied upon the contact between the graphene oxide and the bacterial cells. Another reason which eliminates the number of sharp edges present on the exposed GO surface was that the bacterial toxicity could be achieved using a Langmuir-Blodgett-deposited GO film. According to the justifications presented by the two references, we agree with the connection shown by Rajapaksha et al [43] because many reasons could be related to the process to adsorb bacterias from water and, we should never limit ourselves to a single hypothesis, as happened with the literature Akhavan and Ghaderi [143].

The reference Shao et al [184] reports an indirect proportionality between copper ions removal and the concentration of the used material, which contrasts with what many kinds of literature such as Henriques et al [167] and Zeng et al [172] reported in their studies on mercury and fluoride removal, respectively. The indirect proportionality showed by Shao et al [184] can be related to the type of material and contaminants to be removed from the water. The same study did not report copper removal behavior in concentrations below 75mg/l, which could better understand the relationship between the material concentration and copper removal from water.

6. Conclusion

Using graphene oxide as adsorbent, biological, organic, inorganic, and radiological contaminants can be removed from water until safe for human consumption concentrations. Different methods used to prepare graphene oxide may have a positive or negative impact on the environment and adsorption capacity. Graphene oxide itself absorbs pollutants from water, but the adsorption capacity is improved when the GO is combined with one or more nanomaterials, forming nanohybrids. It was noted that the support membrane's presence also increases the adsorption capacity of contaminants that GO has. Factors such as pH, adsorption time, and concentration of adsorbent material play a significant role in removing different pollutants. Different combinations of GO with other nanomaterials showed a higher capacity of adsorption of contaminants at different pH, time, and concentrations. In implementing water treatment projects using GO, it is essential to understand all factors that can allow us to achieve maximum adsorption of pollutants.

Conflict of Interest

The authors declared that they have no conflicts of interest to this work.

Aknowledgments

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