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# Graphene: A future science material for water treatment

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## REVIEW ARTICLE

## ABSTRACT



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## 1. Introduction

Water, a universal solvent is a very crucial compound in maintaining the ecosystem on the Earth, that is why it cannot be ignored. Hence, if water is called the river of life, it would not be wrong. The percentage of water in the human body (75%) and some plants (95%) also makes it a more desirable compound [1]. The quality of the resources of water is decreasing day by day. The reason behind it is the current changes occurring in agriculture, civilization, environment and geology [2]. The most important and notable point is the number of people (1.1 billion) having no access to safe drinking water [3]. Another thinkable point is the number of people (2.6 billion) having no proper sanitation [3]. Besides, the people having no access to safe sources of drinking water in rural areas are four out of five residents of the world's total people [4]. It is believed that industries and municipalities use approximately 30% of the renewable pure water available globally [5]. This gives rise to a large volume of wastewater containing many chemicals in variable concentrations [6,7]. Some countries are still fighting with such problems. The elimination of the microscopic contaminations/pollutants of wastewater is insufficient [8].

On a worldwide scale, 1.6 million deaths per year are due to controlled access to safe water [9]. In sub-Saharan African countries, nearly 50% of child deaths occur because of drinking contaminated water [10]. According to the current studies,

Graphene with its versatility, is playing the most important role in many fields of science continuously. The most significant role of graphene is noted in the treatment of water. In the present scenario, the supply of safe and pure water has become the main priority. Especially, the denser and more populated areas are demanding of it. Although different materials have been used in water treatment, no material showed the result as good as graphene. The current article deliberates not only the main properties of graphene but also its importance in the treatment of water. Besides, the current review also pronounces the method of graphene separation after the use, i.e. recycling. Efforts are made to discuss the role of graphene-based materials in the treatment of water. Henceforward, this article will be very helpful for researchers, academicians, and administration authorities who are planning and developing new strategies for the removal of pollutants from polluted water.

gastrointestinal diseases (about 15-30%) and health-related problems (6.11%) are because of drinking contaminated water. Contaminated water causes diarrheal diseases easily; it also occurs because of a lack of hygiene and sanitation [11]. The reports of WHO about infectious diseases clearly show that waterborne diseases have been placed at the top of the list of epidemics in 132 countries [12]. The next most frequent disease caused by drinking contaminated water is cholera, followed by typhoid fever, legionellosis, and acute diarrhea [10,13]. All these points confirm that the world is heading towards water scarcity in the future when we will have limited water resources. In such circumstances, the only option to survive is the treatment of water and recycling. Many methods have been developed for water treatment [14,15].

Of course, every technique used for water treatment requires some important materials which have been engaged in various techniques starting from classical ones (obtained from waste and industrial products) to nanomaterials. Recently, a new generation material called graphene came into existence for water treatment [16]. During the literature survey and writing this article, it was observed that graphene has been exploited for water treatment [17]. Also, research is in progress to prepare more advanced graphene materials for water treatment in various techniques. It was also realized that graphene-based materials have not been fully exploited for water treatment and there is a need to do more research in this direction. During the write-up of this article, it was observed

**Table 1.** The characteristics features of single-layer graphene [60].

Features	Values
Density	0.77 mg/m <sup>3</sup>
Young's modulus	1100 GPa
Theoretical BET specific surface area	2630 m <sup>2</sup> /g
Thermal conductivity	5000 W/m.K
C-C bond length thickness	14.2×10 <sup>-2</sup> nm
Resistivity	10 <sup>-6</sup> Ω cm
Density carrier	10 <sup>12</sup> 1/cm <sup>2</sup>
Fractural strength	125 GPa
Optical transparency	97 %
Electron mobility	200000 cm <sup>2</sup> /V.s

that systematic research is still needed on water treatment using graphene. Therefore, the current paper with its novelty describes the developments based on graphene for water treatment.

## 2. Graphene as a promising material

Graphene-based materials have played an important role in water treatment [18] during the last few years. Some people have used graphene for the treatment of water in different approaches and consequently, a few reviews are available in the literature on water treatment using graphene [19-21]. It was recognized that most of the work on water treatment using graphene and its compounds, is on adsorption techniques with only a few papers on membranes, and other methods [16,22]. Therefore, it was tried to find out why the researchers are not attracted to the applications of graphene and its compounds in membrane technology and other methods. In addition, the suggestions to search the options for this wonderful material regarding poisonousness and hazards are given.

## 3. Materials based on graphene

If the description of graphene is needed, we have to start with a crystalline carbon material, i.e. graphite which exists in the form of similar fixed layers [23]. Each layer has many *sp*<sup>2</sup> hybridized carbon atoms surrounded by other three *sp*<sup>2</sup> hybridized carbon atoms. In the hexagonal array, the detachment between the atoms of carbon is 1.42 Å [23]. The covalent bonding is the intra-planar force among the layers. It is greater than van der Waal's force [23]. The intercalation compounds of graphite may be formed by reacting with a variety of chemicals. These properties may be used to modify graphite's physical and chemical properties including surface substitution and modification. Graphite intercalation compounds have unique features to be exploited in water treatment due to their excellent sorption properties for contaminants variety [24].

With a wide range of properties, graphene is also considered a marvelous material [25]. It is being used in a variety of fields including water treatment. An adhesive tape was used to prepare a single layer of graphene from single-crystal graphite [26]. Later on, a single layer of graphene was obtained by developing more advanced methods [27]. Generally, plane and single layers of graphene are steadily over substrates. On the other hand, graphene layers are not stable and prone to mold to a roll form. These sole layers may be stabilized by reacting them in organic and aqueous media [28,29]. Normally, the structure of graphene remains deformed due to the attachment of some atoms and chemicals. It is the concept of everyone that graphene is a single layer but, nowadays, a bundle of 10 layers may be considered graphene [30]. According to Ahmadi-Moghadam and Taheri [31], a bundle of 10 or more layers may be called a graphite nanosheet or exfoliated graphite nanoplatelet. The properties of graphene depend on the number of layers. Graphite with its cellular structure may be obtainable with more than 50 nm thickness [32].

In a single sheet of graphene, a vacant *p*-orbital is observed because of the presence of *sp*<sup>2</sup> hybridized carbon atoms [33]. The thermal conductivity of the suspension of graphene was found ~5000 W/mK [34]. The surface area of graphene specified theoretically is 2630 m<sup>2</sup>/g [35]. A drastic decrease of ~2000 W/mK in the conductance was observed when graphene is in interaction with the support or even slanted with additional sheets of graphene [36]. The range of thickness of graphene was found from 10 to 100 nm because of its reasonable thermal conductivity ( $\kappa \approx 1500$  W/mK) [37]. Graphite may be easily functionalized and integrated with a variety of ways [38] because of its tunable and flexible carbon skeleton.

Distinctive structural features [39], miniature thickness [40], good tensile strength, impenetrability [41-45] and a good mechanical strength [46], make this material marvelous in water treatment. The graphene may be made in the form of a membrane by creating sub-nanometer pores with the help of a beam of electron-ion, the bombardment of cluster/ion, doping, oxidation, etc. [47-50]. Such types of pores may be used to discover the transportation of gaseous molecules and ions [51]. Few layers of graphene (FLG), nanosheets of graphene (GNS), oxide of graphene (GO), nanoribbons of graphene, and reduced graphene oxide (rGO) are considered graphene derivatives.

Electrochemical reduction, decomposition (microwave, thermal, photochemical), exfoliation, etc. [52-54] are methods for the preparation of graphene sheets and their nanocomposite materials. The separation of a single graphene layer of graphite has attained great care owing to outstanding physico-chemical features such as good Yong's modulus value, electrical and thermal conductivity, specific surface area and fracture strength [55]. Due to these facts, graphene nanosheets amazed scientists for widespread applications in science and technology [25]. There are two types of nanoplatelets *i.e.*, only a few layered (1-10 layers) and more layered (10-60 layers). Figure 1 shows some nanomaterial-based structures of graphene. Figure 2 shows the representation of the synthesis of graphene nanocomposite materials. Interestingly, Figure 3 has the surface-modified supra-molecular structure of graphene. Figure 4 shows the supra-molecular structure of the oxide of graphene.

## 4. Oxides of graphene

The variation in the properties of the graphene oxide (GO) has been observed with the change in pHs as well as the ionic balance of water [57]. Besides, the mechanism responsible for the surface activity was also described by using experimental and simulation studies at various pHs [57]. It was observed that -COOH groups protonated with the sheets of GO in acidic conditions; leading to agglomeration [58]. A GO-water-GO roll structure was observed by a simulation study [59]. Contrarily, at high pH, carboxyl groups were found hydrophilic. The individual sheets of GO showed the capacity to dissolve water-loving salt. Table 1 is given to show the observed properties of single-layer graphene.

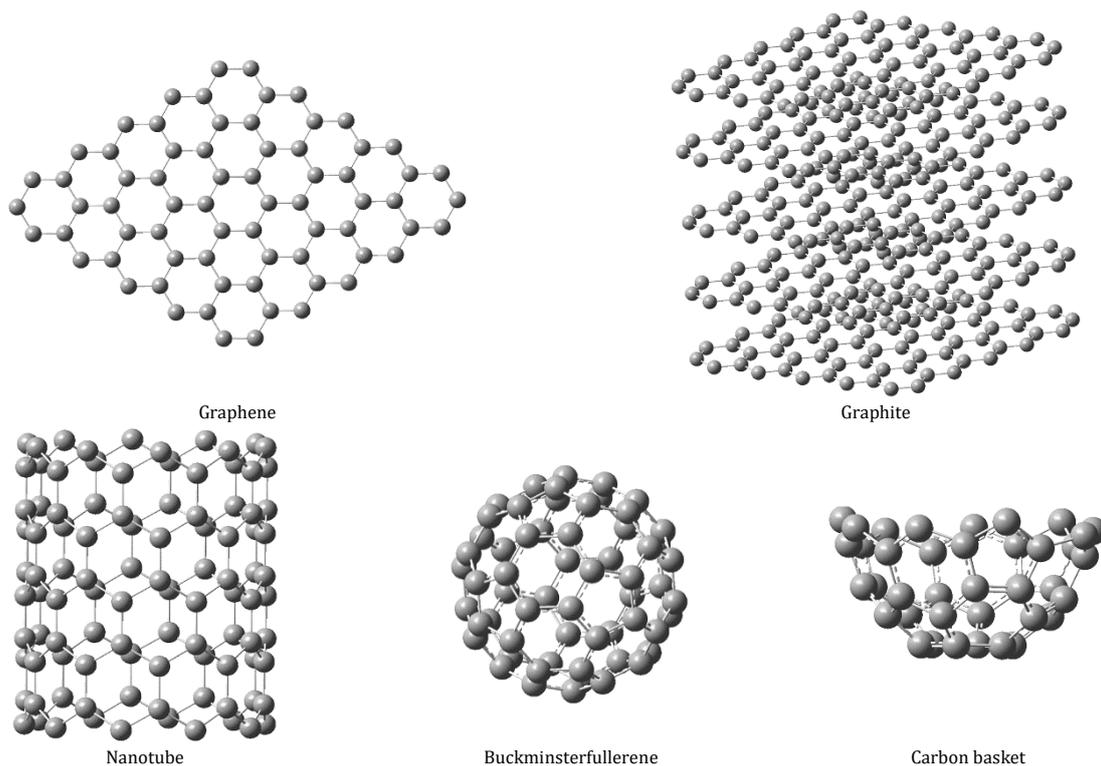


Figure 1. Some carbon-based nanomaterials.

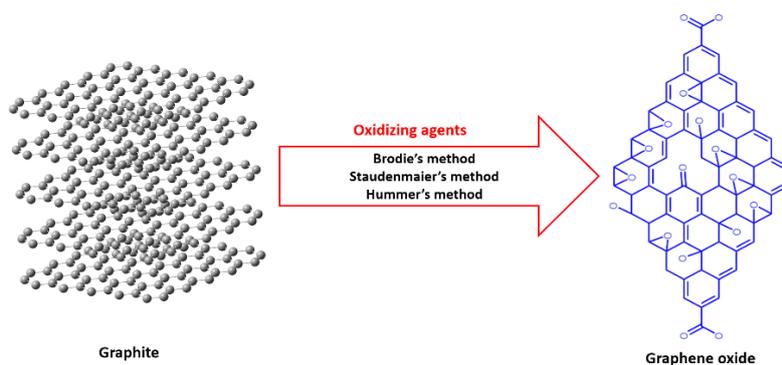


Figure 2. Representation of graphene oxide material synthesis [56].

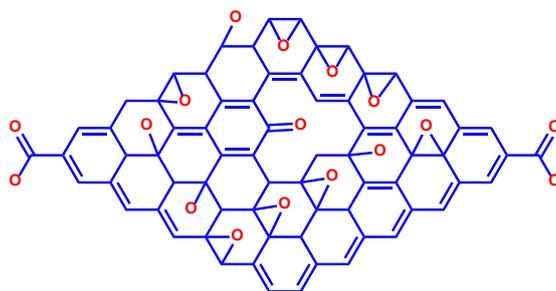


Figure 3. Graphene with a modified surface.

The effects of various concentrations of salts (magnesium chloride, sodium chloride, calcium chloride) and pH were studied by Chowdhury *et al.* [61] on GO stability. It was observed that ionic strength and the type of salt showed a significant effect on GO steadiness because of electrical double layer compression. It was observed that calcium chloride weakened GO more than magnesium and sodium chloride. It

was because of the robust binding ability of calcium ions with -OH and -CO functional groups of graphene oxide. The critical coagulation concentration (CCC) of GO was 44 mM NaCl, 0.9 mM CaCl<sub>2</sub> and 1.3 mM MgCl<sub>2</sub> [61]. The accumulation and firmness of GO in an aqueous solution trailed the colloidal theory (DLVO

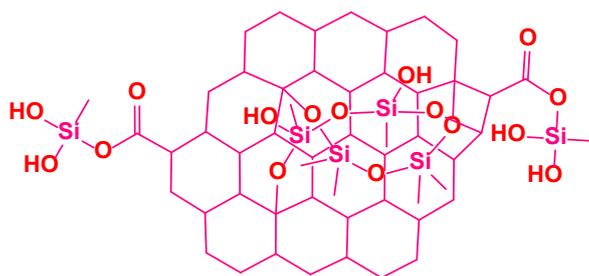


Figure 4. Graphene oxide with supramolecular structure.

and Schulze-Hardy rule) [62]. However, in shape, GO had no similarity to a sphere. The organic matter considerably enhanced the strength of oxide of graphene in water chiefly because of steric repulsion. The steadiness studies showed that GO was extremely stable in laboratory, synthetic, surface and natural waters. In addition, GO was rapidly settled down in laboratory synthetic water but it continued constant in contaminated water [61]. In addition to this, as per the author, GO was constant in the aquatic system of nature. Ren *et al.* [63] reported the stabilization of GO in solutions of salts like  $\text{CaCl}_2$  and  $\text{MgCl}_2$ . It was due to their effective charge selection and neutralization. Additionally, polyacrylic acid and  $\text{NaH}_2\text{PO}_4$  enhanced the firmness of the oxide of graphene with the rise in basic nature because of steric repulsion and electrostatic forces [63]. In water treatment, the exploitation of this information may be used for the prediction of the possible use of GO.

## 5. Graphene role in water treatment

Of course, from various points of view, water treatment is an essential issue [64,65]. For a few years, graphene and its materials are playing an important role in water treatment through adsorption, photo-degradation, and filtration methods [16,66]. This is due to the various functional groups and flexibility of operation [67]. Graphene and its composites have been used in many forms in water treatment. Some of its applications in water treatment are given here.

### 5.1. As a degrader of pollutants

The photocatalytic degradation of organic pollutants is also gaining a position in water treatment [68]. Some graphene-based materials have also been tested by some researchers [69-75]. The graphene-based materials are attracting researchers because of the mobility of their large surface area, charge carriers, transparency, elastic structure, and satisfactory thermal and electrical conduction [76]. Good catalytic performance depends on the synergic effect of the enlarged specific surface area of graphene nanosheets, and the ionic interaction among organic pollutants and oxygenated functional groups of the graphene materials [77]. The photocatalytic degradation is enhanced by providing adsorption surface area, the acceptor of electrons, and a photosensitizer to quicken decomposition occurring photolytically [78]. Some reviews are available on the photocatalytic degradation of pollutants by graphene-based materials [79-82]. These discuss the synthesis, applications, performance, and mechanism of water purification.

Chen *et al.* [69] reported the elimination of methyl orange dye by  $\text{TiO}_2$ /chitosan/RGO macroporous composite. The authors described the synthesized material as low cost with good performance. The photocatalytic removal of medicinal pollutants was observed by Lin *et al.* [83] using the nanocomposites of  $\text{TiO}_2$ -reduced oxide of graphene. The pollutants were degraded in the presence of UV and visible light. A good performance was because of 2.85 eV as bandgap

energy [84]. Therefore,  $\text{TiO}_2$ -RGO showed more efficiency under UV irradiation because of the reduced recombination rate of photoexcited electrons-hole pairs. The effect of diverse ZnO morphologies was reported by Kang *et al.* [85] on the photocatalytic activity of nanocomposites of ZnO/RGO; with the separation efficiency of the photoinduced charges, photo-absorption, and photocatalytic concert of the composite of ZnO/RGO. The photodegradation of raw textile wastewater and reactive Black 5 was reported by Silva *et al.* [86] using heterogeneous photo-Fenton reaction as well as amino- $\text{Fe}_3\text{O}_4$ -functionalized oxide of graphene as a nanocatalyst. The authors carried out kinetics studies along with the applications of Chan and Chu model [87]. The experiments based on photolytical degradation showed subtraction efficacies of 53.25% for apparent color and 64.55% for turbidity. Luna-Sanguino *et al.* [88] prepared  $\text{TiO}_2$ -RGO nanocomposites by hydrothermal method for the photodegradation of alachlor, pyrimethanil, methomyl and isoproturon pesticides. Nanocomposite material based on graphite was synthesized by Arshad *et al.* [89] and many usual techniques (SEM, FTIR, Raman, and UV-Visible Spectroscopy) were used to characterize them. Methylene blue was photo-degraded using the developed materials. Different titania composites based on graphene were synthesized by Liu *et al.* [90] using the sol-gel method, and many usual techniques (XRD, FT-IR, DRS, UV-Vis., TGA-DTA and SEM) were used to characterize them. Methylene blue was photo-degraded using the developed materials. Modified solid graphite electrodes were synthesized by Malinauskas *et al.* [91] using adsorption and a droplet evaporation method, and many usual techniques (XRD, FT-IR, DRS, UV-Vis., TGA-DTA and SEM) were used to characterize them. Methylene blue was photo-degraded using the developed materials. Solid graphite electrodes were modified with the redox dyes Nile blue toluidine, while the determination of dyes adsorption was done using cyclic voltammetry. As per the authors, good photodegradation of 200  $\mu\text{g/L}$  of each pesticide was observed. Table 2 is given for the literature data based on the photodegradation of organic pollutants.

### 5.2. Graphene as an adsorbent

The removal of organic pollutants by the adsorption method is being used to treat wastewater [10,101-103]. Thousands of papers are available on this method of water treatment [133-137]. This method is gaining popularity due to the easiness of its use, the inclusion of source reduction, recovery of the pollutants for future beneficial applications, etc. Despite thousands of publications, adsorption could not be a good place to treat wastewater on a large scale because of the non-availability of good adsorbents. However, researchers are exploring the hope of a good adsorption capacity of graphene and its derivatives. Graphene is gaining importance in adsorption due to its large surface area [138]. Besides, graphene

**Table 2.** Removal of pollutants by using graphene-based materials.

Methods	Pollutants class	Pollutants name	Materials	Experimental conditions	Capacity	Refs.		
Pd	OP	Methyl orange	TiO <sub>2</sub> /Chitosan/Reduced graphene oxide	1.0wt % RGO, UV lamp ( $\lambda = 365 \text{ nm}$ )	97%	[69]		
		Methyl orange	Graphene-ZnO (GZ)	UV light and sunlight	97.1%	[70]		
		Rhodamine B	Copper Phthalocyanine (CuPc) functionalized reduced graphene oxide	Visible light	28%	[71]		
		Rhodamine B	Reduced graphene oxide-BiPO <sub>4</sub>	UV light	87.5%	[72]		
		Congo Red	CuS-Reduced graphene oxide	Sunlight	98.76%	[73]		
		Methyl orange	Ag-Reduced graphene oxide	Visible light	99.71%	[74]		
		Rhodamine B	dS/Reduced graphene oxide	Visible light	93%	[75]		
		Methyl orange	3D CeVO <sub>4</sub> /Graphene aerogels	Visible light	98%	[92]		
		Methylene blue, methyl orange, and rhodamine B	CdS-Graphene	Visible light	95%	[93]		
		Methylene blue	Polythiophene (PTh)-Reduced graphene oxide-TiO <sub>2</sub>	Visible light	14.17%	[94]		
		Acid blue 74	Ag-Ag <sub>2</sub> O-ZnO/GO	Visible light	90%	[95]		
		Cyclohexane	GO-Ag	Solar light	94%	[96]		
		Tetracycline hydrochloride	Ag loaded TiO <sub>2</sub> nanowire arrays/GO coated carbon fiber	Visible light	95.73%	[97]		
		Enrofloxacin	Polyacrylic acid grafted carboxylic graphene/titanium nanotube	Sunlight	99.0%	[98]		
		Chloramphenicol	Ce(MoO <sub>4</sub> ) <sub>2</sub> nanocubes/GO	Visible light	99%	[99]		
		Tetracycline	Reduced graphene oxide-Cu <sub>2</sub> O/Bi <sub>2</sub> O <sub>3</sub>	Visible light	75%	[100]		
		Ad	IP	Lead (+2)	Graphene	40 mg/L (pH=4.0) at 303 K for 15 h	22.42 mg/g	[101]
				Lead (+2)	Graphene	40 mg/L (pH=4.0) at 303 K for 15 h	35.21 mg/g	[101]
				Arsenic (+3)	Graphene/Mn <sup>2+</sup> Fe <sup>3+</sup> O <sup>2-</sup>	1-8 mg/L (pH=7.0 $\pm$ 0.1) at 300 $\pm$ 1 K for 2.5 h	14.42 mg/g	[101]
				Antimony (+3)	Graphene	1-10 mg/L (pH=11.0) 303 K for 4 h	10.919 mg/g	[102]
Lead (+2)	Graphene			40 mg/L (pH=4.0) at 303 K for 15 h	35.46 mg/g	[103]		
Lead (+2)	Graphene oxide			5-300 mg/L (pH=6.8) at 298 $\pm$ 2 K for 24 h	367 mg/g	[104]		
Lead (+2)	Graphene oxide			5-300 mg/L (pH=7.0 $\pm$ 0.5) at 298 $\pm$ 5 K for 24 h	692.66 mg/g	[105]		
Lead (+2)	Poly(N-vinylcarbazole)-graphene oxide			5-300 mg/L (pH=7.0 $\pm$ 0.5) at 298 $\pm$ 5 K for 24 h	982.86 mg/g	[105]		
Copper (+2)	Graphene oxide aerogel			50-75 mg/L (pH=6.3) at 283 K for 0.5 h	17.73 mg/g	[106]		
Lead (+2)	EDTA modified graphene oxide			5-300 mg/L (pH=6.8) at 298 $\pm$ 2 K for 24 h	525 mg/g	[107]		
Chromium (+6)	Graphene/Fe			25-125 mg/L (pH=4.25) at 293 K for 4 h	162 mg/g	[108]		
Chromium (+6)	Graphene /Fe@Fe <sub>2</sub> O <sub>3</sub> @Si\SiO <sub>2</sub>			1 g/L (pH=7.0)	1.03 mg/g	[109]		
Lead (+2)	SiO <sub>2</sub> /Graphene			20 mg/L (pH=6.0) at 298 K for 1 h	113.6 mg/g	[110]		
Arsenic (+5)	Double hydroxides			0.5-20 mg/L (pH=4.0-9.0) at room temp. for 24 h	23.78 mg/g	[111]		
Copper (+2)	Graphene oxide/Ferric hydroxide hydrogel			0-120 mg/L (pH=5.1) at 294 $\pm$ 1 K for 10 h	70 mg/g	[112]		
Arsenic (+5)	Graphene oxide-ZrO(OH) <sub>2</sub>			2-80 mg/L (pH=7.0 $\pm$ 0.2) at 298.5 $\pm$ 0.2 K for 0.25 h	84.89 mg/g	[113]		
Arsenic (+3)	Graphene oxide-ZrO(OH) <sub>2</sub>			2-80 mg/L (pH=7.0 $\pm$ 0.2) at 298.5 $\pm$ 0.2 K for 0.25 h	95.15 mg/g	[113]		
Copper (+2)	Sulfonated magnetic graphene oxide composite			73.71 mg/L (pH=5.0) at 283.15 K for 6 h	50.678 mg/g	[114]		
Mercury (+2)	Polypyrrole-Reduced graphene oxide			50-250 mg/L (pH=3.0) at 293 K for 3 h	979.54 mg/g	[115]		
Arsenic (+3)	Magnetite-Reduced graphene oxide			3-7 mg/L (pH=7.0) at 293 K for 2 h	10.20 mg/g	[116]		
Arsenic (+3)	Reduced graphene oxide-Fe <sub>3</sub> O <sub>4</sub>			2-6 mg/L (pH=7.0) at 298 K for 1 h	21.2 mg/g	[117]		
Arsenic (+3)	Reduced graphene oxide-Fe(0)			2-6 mg/L (pH=7.0) at 298 K for 1 h	37.3 mg/g	[117]		
Arsenic (+3)	Reduced graphene oxide-Fe(0)/Fe <sub>3</sub> O <sub>4</sub>			2-6 mg/L (pH=7.0) at 298 K for 1 h	44.4 mg/g	[117]		
Mercury (+2)	Reduced graphene oxide-Fe(0)/Fe <sub>3</sub> O <sub>4</sub>			2-6 mg/L (pH=7.0) at 298 K for 1 h	22.0 mg/g	[117]		
Lead (+2)	Reduced graphene oxide-Fe(0)/Fe <sub>3</sub> O <sub>4</sub>			2-6 mg/L (pH=7.0) at 298 K for 1 h	19.7 mg/g	[117]		
Chromium (+6)	Reduced graphene oxide-Fe(0)/Fe <sub>3</sub> O <sub>4</sub>			2-6 mg/L (pH=7.0) at 298 K for 1 h	31.1 mg/g	[117]		
Arsenic (+3)	Reduced graphene oxide-Fe(0)			2-6 mg/L (pH=7.0) at 298 K for 1 h	37.3 mg/g	[117]		
Cadmium (+2)	Reduced graphene oxide-Fe(0)/Fe <sub>3</sub> O <sub>4</sub>			2-6 mg/L (pH=7.0) at 298 K for 1 h	1.91 mg/g	[117]		
Lead (+2)	Reduced graphene oxide-iron oxide			10-15 mg/L (pH=6.5 $\pm$ 0.1) at 303 K for 48 h	454.55 mg/g	[118]		

Table 2. (Continued).

Methods	Pollutants class	Pollutants name	Materials	Experimental conditions	Capacity	Refs.
	OP	Crystal violet	Graphene oxide	0.25–1.0 mg/L (pH = 2-10) at 298–313 K for 2 h	22.90–24.93	[119]
		Methylene blue	Graphene oxide	400 mg/L (pH = 6-8) at 298 K for 24 h	581.56 mg/g	[120]
		Naphthalene	Graphene nanoplatelet/MIL-101	30 mg/L (pH = 7) at 298 K for 10 min	075 g	[121]
		1-Naphthol	Sulfonated graphene nanosheets	~6.4 mmol/g at 333.15 K for 10-15 h	~2.3-6.4 mmol/g	[122]
		Naphthalene and 1-naphthol	Modified graphene nanosheets	(for naphthalene) = 0.04 g/L, (for 1-naphthol) = 0.08 g/L (pH = 7) 120 °C for overnight	2.3-2.4 mM/g	[123]
		Bisphenol	Graphite nanoplatelets	1-10 mg/L (pH = 5.5) at 298 K for 30 min	850mg/g	[124]
		Bisphenol A	Exfoliated graphene nanoplatelets	0.5 mg/L (pH = 5.5) at 298 K for 30 min	850 mg/g	[124]
		As(III)	Graphene nanoplatelet	0.2 g/L (pH=7) at 298 K for 2 h	11.34 mg/g	[125]
		Methylene blue or malachite green	Graphite oxide	Con. 1.0 mg/mL at 298 K for 40-60 min	3.0 & 6.0 mg/g	[126]
		Methylene blue	Graphene	10 mg/L (pH = 6.5) at 298 K for 600 min	153.85 mg/g	[127]
		Methyl violet from	Nano-graphite/Fe <sub>3</sub> O <sub>4</sub> composite	0.5 × 10 <sup>-2</sup> mg/mL (pH = 4) at 298 K for 10 min.	41-48 mg/g	[128]
		Safranin dye	Graphite oxide	0.5 g/L (pH = 6) at 298 K for 45-60 min	97.78%	[129]
		Ibuprofen	Graphite oxide	1 g/L (pH = 7) at 298 K for 60 min	98.17%	[130]
		Crystal violet	Graphite oxide	100 mg/L (pH = 8) at 298 K	43.38 mg/g	[131]
		Bisphenol A	Graphene	10 mg/L (pH = 2-7) at 298 K for 2 h	182.0 mg/g	[132]

\* Pd: Photo-degradation; Ad: Adsorption; OP: Organic pollutants; IP: Inorganic pollutants.

derivatives show a fast and large adsorption capacity with respectable constancies [139]. Graphene and its nanomaterials have been a perfect adsorbent for the removal of the pollutants present in contaminated water [101-104,134-137]. Magnetic graphene oxide (MGO) was synthesized by Ain *et al.* [140] and many usual techniques (UV-VIS, FT-IR, SEM, XRD, and VSM) were used to characterize them. Pb<sup>2+</sup>, Cr<sup>3+</sup>, Cu<sup>2+</sup>, Zn<sup>2+</sup>, and Ni<sup>2+</sup> were removed from water using the synthesized adsorbent. Many kinetic models were used to fit data well. The various parameters such as initial concentration, contact time and pH were also optimized. The maximum adsorption capacities at pH = 5.0 for Pb (II), pH = 6 for Cr (III), pH = 6 for Cu (II), pH = 7 for Zn (II) and pH = 8 for Ni (II), ions were 200, 24.330, 62.893, 63.694 and 51.020 mg/g, respectively [141]. A novel adsorbent GO-TETA-DAC was synthesized by Yao *et al.* [142] and many usual techniques (FT-IR, BET, and SEM) were used to characterize them. Cu (II) and Pb (II) were removed from water using the synthesized adsorbent. Many kinetic models were used to fit data well. The various parameters such as initial concentration, contact time and pH were also optimized. The re-dispersion of GO paper was involved in the synthesis of a new adsorbent (GO/CH paper) by Cortinez *et al.* [143] and many usual techniques (FT-IR and XRD) were used to characterize them. Methylene blue and methyl orange were removed from water using the synthesized adsorbent. Many kinetic models were used to fit data well. In addition, the optimization of contact time, pH and initial concentration was done.

Graphene and its composites are used in water treatment for the last few years [104,135,136]. Banerjee *et al.* [129,130] did some work on the adsorption of pollutants using graphene in 2015 and 2016. In 2015, the authors did a simulation study for the removal of azo dye from wastewater using graphene oxide nanoplatelets as an adsorbent. Later on, this adsorbent was prepared and identified by SEM, FTIR, AFM, and TEM. This material was used to remove safranin dye in wastewater with a reported capacity of 97.78%. In 2016, the authors used the same material for the removal of ibuprofen. The authors described this material as efficient on an industrial scale. Gan *et al.* [60] described the interactions among pollutants and the adsorbent during the removal of polycyclic aromatic hydro-

carbons from a coking plant soil. The authors reported  $\pi$ - $\pi$ , H- $\pi$  and anion- $\pi$  interactions among aromatic hydrocarbons and nano-sulfonated graphene. A comparative study between GO and GO-COOH nanoparticles for the absorption of Cu(II) was also performed by White *et al.* [144]. The simple batch-adsorption mode was used for it. It was found that the adsorption capacity of GO-COOH was greater than that of GO as per Langmuir isotherms. Besides, the reusability of both adsorbents was also estimated. A brief glimpse regarding the removal of organic pollutants by using graphene materials is given in Table 2.

### 5.3 Graphene as a filter

Because of having many pores, graphene and its nanosheets are also used in the filtration method of water treatment [145]. Of course, there are other polymeric membranes but the most important thing to be noted is the requirement for less energy [146]. Nanosheets based on graphene display tremendous Physico-chemical properties for the treatment of water through filtration. Graphene-based membranes displayed better mechanical strengths, and the capability to work below higher pressure and under other circumstances [147,148]. Nanosheets based on graphene oxide display tremendous antifouling capability; an asset compulsory in the treatment of water through filtration [149]. Nanosheets based on graphene oxide display efficacy in only the flow of water by preventing other constituents [150]. Since 2009, the main use of graphene is the preparation of the membrane for water treatment with the main stress on synthetic nanosheets having different sizes of pores; a variety fit for purification [48,51]. The importance of graphene-based membranes involves nano-filtration and osmosis in the reverse direction for seawater purification [147,151], the passage of selective ions [146,152] and the separation of gases [153]. The bioreactor-based membrane has appeared and fascinated scientists in the treatment of wastewater. The membrane fouling problem is the main trouble in the case of the membrane bioreactors, which complications maximum efficacy consequential to small membrane life and raised effective budgets [154]. The applications of graphene-

based material were reviewed by Manawi *et al.* [155]. The authors defined the treatment of water using the materials cited above. The presented review was clearly showing the bright future of graphene in the treatment of water.

Of course, graphene oxide gives better results as compared to pure graphene in the water filtration process [73-75]. Generally, the diameter of pores in graphene oxide is found approximately 5.0 Å [156], which is prolonged to 9 Å [157] on hydration, consequently, the molecules with the size of 4.5 Å [158] can also pass, while molecules with a size greater than this are prevented [159]. There are many methods for the preparation of nanosheets based on graphene oxide such as layer-by-layer deposition, vacuum filtration, spin coating, drop-casting, *etc.* The pores of graphene oxide can be modified by the functionalization with either H atoms or -OH groups. Obviously, different effects are produced on the filtration of water [160]. Rather than hydrogenated pores, hydroxylated pores show better results in the increment of the permeability of water [145]. The reason behind it was the increase in water flux. Greater water flux through the hydroxylated pores; as compared to hydrogenated pores; is due to the higher cross-sectional part presented for the molecules of water to pass. Hydrogenated holes consume about a 25% decrease in surface area as compared to hydroxylated holes. The diverse methods have been approved to include graphene oxide in polymer-forming solutions; either during the manufacture of the membrane or post-coating of the assembled membrane. Thus, mechanical strength, permeability, and antimicrobial properties [161,162] were made better.

Polyacrylamide/graphene-based nanocomposites were synthesized by Manafi *et al.* [163] and many usual techniques were used to characterize them. An electrodes filter based on functionalized graphite nanoplatelets was used [164], and electron microscopy was used to characterize it. Na(I), As(III) and As(V) were removed from seawater using the membrane. Many kinetic models were used to fit data well. The various parameters such as initial concentration, contact time and pH were also optimized. The maximum adsorption capabilities for As (V), As (III) and Na metal ions from seawater, were 27.0, 29.0 and 32.0 mg/g respectively. Moreover, the nanoplatelets of exfoliated graphite were used by Crock *et al.* [165]. The removal of methyl orange dye from the water was done by Filice *et al.* [166] using graphene oxide. Many kinetic models were used to fit data well. The various parameters such as initial concentration, contact time and pH were also optimized. Lee *et al.* [161] described that a new generation membrane having good anti-fouling ability can be obtained by the addition of only 1.0 wt% of graphene oxide in the preparation of the membrane. After that, water treatment was done using the established membrane. Hosseini *et al.* [167] used the nanoporous graphene oxide (NPGO)-based membranes for the removal of unwanted impurities from water. The surface and edge of holes played a significant role in water flux as per the results obtained. In the rejection of salt, a high efficacy with high performance in water flux was shown by the membrane based on NPGO [167]. The NPGO acted as an ultra-permeable membrane because of the existence of the hydrophilic groups on the surface, and the gotten water flux for NPGO was around 77% superior as compared to graphene.

## 6. Recovery of graphene materials

The most tedious job after the water treatment is the regaining of graphene or its modified nanocomposites. Essentially, expert hands with careful management are required for the departure of material based on graphene. Mostly, the nature of graphene also matters in the recovery of graphene-based material from water [168]. For the recovery of nanoparticles/graphene oxide-based materials from water, various methods have been used and applied [169]. Examples, cross-flow

filtration, filled flow fractionation, magnetism, continuously filled flow fractionation, ultra-centrifugation, ultra-filtration and electric field [107]. Of course, magnetic properties are displayed by magnetic graphene materials. Due to this reason, it can be recovered on a high gradient magnetic field. More complex separation can also be done using this method as well as using functionalized magnetic graphene [107]. Magnetic properties, magnetic field gradients and size play a key role in the gradient magnetic separation [168]. Besides, the recovery of nanoparticles has also been done using a cross-flow membrane [169]. Permeate convection, Brownian diffusion and electrokinetic migration were used to control the nanoparticle transport in cross-flow membrane filtration. Besides, a little bit of contribution of shear-induced diffusion, gravitational settling, axial convection and inertial lift is also involved [107]. Mainly, the size and the type of the membrane are involved in this technique. The nanoparticle size determines the selectivity of the membrane [170,171]. Graphene-based materials can be recovered by the centrifugation process, which can be actual due to their high efficacy, and the ability of accessible and free of accumulation. For such constituents, another method i.e. the velocity-based rate zonal and density-related isopycnic centrifugation may be operative. Similarly, for graphene, another type of centrifugation i.e. the isopycnic centrifugation may be an advantage [172].

As per literature observation, solvents depending on pH play a key role in the recovery process of adsorbents [173]. In the case of materials based on graphene, one article was found [60] that labels the recycling of graphene. It described the reusing of graphene-based nano-sulfonation, after ex-situ soil washing for polycyclic aromatic hydrocarbons from a coking plant soil. The decrease in removal on subsequent regeneration cycles was reported by authors [60]. One of the most important characteristics is considered the management of the used graphene. The paper regarding the management of the used graphene was not found yet, while everybody is aware. Therefore, there are many resources such as cement, steel, and brick [174] where recycled graphene may be used.

## 7. Future perspectives

Significantly, a large number of articles describing the role of graphene-based substances, exist [101-104,134-137]. Of course, the time would come when graphene-based materials in water treatment will replace many other existing classical materials [175]. Their unique features are the ideal products for using these materials in adsorption and membrane processes of water treatment. Therefore, it seems that in future water treatment, recycling and reuse will be an easy job with economic considerations. Despite a remarkable and thrilling future, some challenges are existing to materialize the utility of graphene-based materials. Some significant hurdles include the economic synthesis of these materials along with proper characterization and proper industrialization [176]. There is a great need to explore the best ways to modify the surface of these materials so that they may have a high capacity to remove pollutants. The other challenges limiting their utilities are ecological and health dangers. Consequently, there is a need to ascertain the complete removal of these materials in water; after treatment [173]. Overall, it was realized that the future is quite bright for graphene-based materials but we have to carry out systematic research to solve real-world problems. Besides, the recycling of materials is one of the most important variables to control the water treatment economy. Therefore, it is a major challenge to recycle used materials.

## 8. Conclusion

Finally, it can be concluded that graphene-based materials have been given great attention in water treatment due to their

unique features. The most important properties making these materials ideal are high surface area, small particle size, free active valences, charge carriers, high mobility, etc. The removal capacity of these materials is very wide-ranging from inorganic to organic water pollutants. Much work has not been done in exploring these materials for water treatment. However, for practical applications in water treatment, the environmental friendly of the whole life cycle, including the synthesis, structural modification, structural assembly and application, should be considered. Cost efficiency is a vital factor to be considered in practical water purification. Large-scale production requires highly efficient synthetic methods, and the fabrication methods should be improved with the goal of environmental protection. The desired method should be recyclable with no toxic emission while also being time-efficient. Most research work is limited to publication and many applications in solving real-world problems have not been recognized. Therefore, there is a great need to do more research to find out the best material and technology for water treatment. Besides, it is also important to explore the utility of these materials to provide safe water at a large scale by developing novel technologies.

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Conflict of interest: The author declares that he has no conflict of interest.  
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#### CRedit authorship contribution statement

Conceptualization: Mohammad Suhail; Methodology: Mohammad Suhail; Software: Mohammad Suhail; Validation: Mohammad Suhail; Formal Analysis: Mohammad Suhail; Investigation: Mohammad Suhail; Resources: Mohammad Suhail; Data Curation: Mohammad Suhail; Writing – Original Draft: Mohammad Suhail; Writing - Review and Editing: Mohammad Suhail; Visualization: Mohammad Suhail; Funding acquisition: Mohammad Suhail; Supervision: Mohammad Suhail; Project Administration: Mohammad Suhail.

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