

GRAPHENE OXIDE-BASED MATERIALS FOR THE TREATMENT OF CONTAMINATED WATER: A REVIEW

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ABSTRACT

Environmental pollution, especially water pollution, has become a serious problem worldwide. Therefore, a number of methods have been investigated and applied to deal with this problem. Using novel materials for environmental treatment is one of the most popular approaches. Among them, graphene oxide (GO) and GO-based materials have attracted much attention due to their unique properties. GO-based materials are able to adsorb various types of contaminants onto their high specific surface areas with the addition of various multifunctional groups. Besides, GO-based materials express both catalyst activity for the degradation of pollutants and antibacterial properties. To synthesize GO-based materials, other components (polymers, metals and metal oxides) are usually combined onto the surface of previously prepared GO. This paper reviews and discusses the synthesis of GO-based materials and their use in treating contaminated water.

Keywords: Adsorption, antimicrobial, contaminant degradation, graphene oxide, GO-based materials.

1. INTRODUCTION

Over the last few decades, industrialization, population growth, and urbanization have resulted in the severe problem of environmental pollution, especially water contamination. A variety of contaminants, including heavy metals, organic compounds, and biofactors in water, have caused adverse impacts on human life (Wang *et al.*, 2013). Therefore, a number of technologies, including physical, biological, and chemical removal processes were developed for contaminant removal from water (Mansoori *et al.*, 2008; Gadd, 2009). Among them, using materials for contaminant adsorption or catalytic degradation show many advantages such as easy operation and effective removal of various types of pollutants (Neyens & Baeyens, 2003; Fu & Wang, 2011). The adsorptive materials are usually highly porous or have a high surface area (De Gisi *et al.*, 2016). While catalytic materials should be cheap, harmless, reusable, and could be attached to different

types of inert materials (Malato *et al.*, 2009). Some materials even express both adsorption and catalyst activity like fly ash (Visa *et al.*, 2018) or carbon-based materials (Sweetman *et al.*, 2017). In the carbon-based group of materials, graphene and graphene oxide have attracted tremendous attention due to their unique properties and various applications (Nguyen, 2016), especially in environmental treatments (Gupta Chatterjee *et al.*, 2015; Ersan *et al.*, 2017; Anand *et al.*, 2018; Sherlala *et al.*, 2018).

Graphene is the mono- or several-layer material consisting of sp²-hybrid carbon atoms that link to other atoms by covalent bonds to form a hexagonal lattice (as shown in Figure 1). The structure of graphene makes its specific surface area extremely high (Geim & Novoselov, 2007). Graphene oxide (GO) is a derivative of graphene that includes groups like -OH or -COOH rising from the planar graphene. These functional groups not only maintain the high surface area of graphene oxide but also provide it with hydrophilicity and biocompatibility.

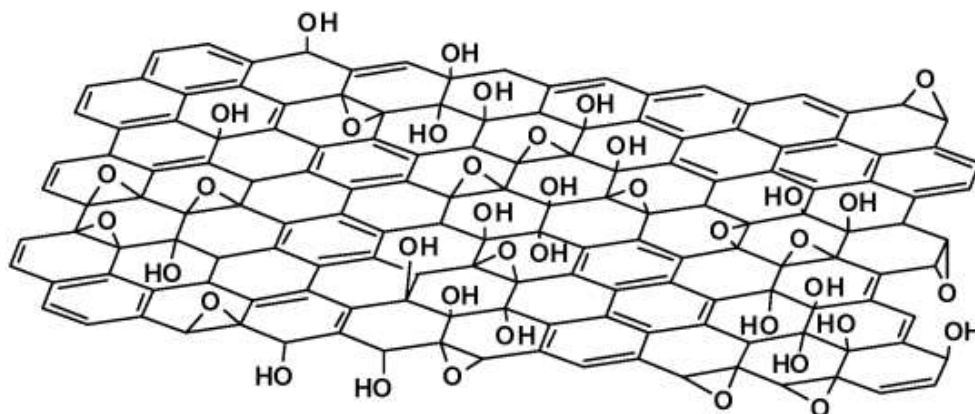


Figure 1. Structure of graphene oxide (adapted from reference He *et al.*, 1998)

The chemistry of GO covers its reduction to reduced GO and reactions of the functional groups (carboxylic acid, hydroxyl or epoxy groups) on the surface of the GO. The resulting products of these reactions are very similar to GO with a variety of possible functions in many applications (Daniel *et al.*, 2010). These applications involving GO materials include being a catalyst for organic chemical reactions (Dreyer *et al.*, 2010), adsorption, and the storage and separation of gases (Gadipelli & Guo, 2015). Especially, thanks to its useful properties, graphene oxide and its derivatives are widely used in the treatment of water, especially for contaminant adsorption and degradation (Kyzas *et al.*, 2014). This review is to overview the synthesis and recent crucial environmental applications of GO-based materials.

2. SYNTHESIS

2.1. Graphene oxide (GO)

It has been reported that GO was first prepared in 1859 (Daniel *et al.*, 2010). However, the Hummers method (Hummers & Offeman, 1958), reported almost a century later, has been used or modified by almost all authors to synthesize GO (Liu *et al.*, 2011; Liao *et al.*, 2012; Liu *et al.*, 2013; Liao *et al.*, 2016). Generally, to obtain GO, graphite is oxidized with strong oxidants. In a typical procedure, 1 g of graphite powder, 0.5 g of NaNO_3 , and concentrated H_2SO_4 are allowed to react with each other at a

temperature below 5°C . Three grams of KMnO_4 is then added slowly to the mixture and followed by adding water and a 2.5 mL solution of H_2O_2 30% (Deng *et al.*, 2013). Modified approaches usually utilize different ratios of oxidants, such as not adding NaNO_3 and adding various amounts of KMnO_4 (Dimiev *et al.*, 2014), or replacing NaNO_3 with other types of oxidants like $\text{K}_2\text{S}_2\text{O}_8$ or P_2O_5 (Gao *et al.*, 2012).

The mechanism of GO formation has been well investigated to be a 3-separate step process. Initially, graphite is converted to the first intermediate called GIC, which is the padding product of the reaction between H_2SO_4 and graphite. In the next steps, GIC undergoes reactions to form pristine graphene oxide (PGO) that in turn reacts with water to become GO in the final step. The second step was proven to be the rate-determining step through XRD and Raman spectroscopy (Dimiev *et al.*, 2014). Based on the starting GO, many other GO derivatives, namely reduced graphene oxide (rGO) and graphene, can be synthesized by the hydrothermal method, electrochemical deposit, or sol-gel technique (Hashim *et al.*, 2016).

2.2. GO-polymer

GO-polymer composites can be obtained by both *in situ* and *ex-situ* incorporation. *In situ* encapsulation of GO into a polymeric matrix during polymerization is an important way to prepare GO-polymer composites in large amounts without the destruction of the

polymeric chain (Hashim *et al.*, 2016). For instance, using aniline as a precursor and in the presence of ammonium persulfate reagent, GO can be grafted at the same time as polyaniline synthesis (Qi *et al.*, 2015). For this process, 200 g of N-Vinylpyrrolidone is combined with a suspension of GO in the presence of N₂ for 30 min while being continuously stirred. Then, the reaction flask is put in an oil bath at 95°C for 10 h. The poly(vinylpyrrolidone) grafted GO is obtained by filtering the reaction mixture through a 0.22 mm-pore-size film (Mauter & Elimelech, 2008). A polypyrrole-GO composite can also be synthesized in a similar process (Zebedius *et al.*, 2015).

In addition, *ex-situ* grafting can be used for the grafting of GO by biopolymers like alginate or chitosan (CS). GO-sodium alginate (SA) hydrogel is synthesized by adding the mixture of GO and SA dropwise to a solution of CaCl₂. When a gel is formed by the cross-linking of the alginate matrix by Ca²⁺ ions, GO is encapsulated into the SA matrix. The GO-SA aerogel is then obtained from the GO-SA hydrogel by lyophilization (Fei *et al.*, 2016). A GO-CS composite can be synthesized in a similar procedure but is not dispersible in aqueous solutions as GO itself and can easily be isolated by filtration or centrifugation (Yang *et al.*, 2013). In another study, GO-cellulose membranes were prepared by combining a GO suspension with urea, sodium hydroxide, and microcrystalline cellulose with the assistance of an ultrasonic bath (Mauter & Elimelech, 2008).

2.3. GO-transition metals and metal compounds

Silver nanoparticles are well known for their antibacterial properties. Combining Ag with GO has been shown to improve the bioactivity of Ag compared to Ag alone. To acquire a GO-Ag composite, GO first has thiol groups attached to its surface. Then, the thiolate GO is mixed with AgNO₃ solutions before reacting with a NaOH solution for 20 h (Vi & Lue, 2016). Ag can also be synthesized *in situ* on the surface of the GO by the reduction of

AgNO₃ in a suspension of GO by HCOONH₄ (Song *et al.*, 2016). In another described process, a 1 mg mL⁻¹ GO suspension, glucose, and PdCl₂ (or HAuCl₄, RhCl₃) were allowed to react for about 30 min before a hydrothermal treatment in a Teflon autoclave at 120°C for 20 h. The prepared sample was washed and then freeze-dried to obtain the noble metal promoted GO (Tang *et al.*, 2010)

The incorporation of GO and transition metal oxides, especially magnetic oxides for water treatment, have attracted much attention due to the combined properties of the composites. Magnetic GO can be easily separated from water by the application of an external magnetic field (Siddiqui & Chaudhry, 2018). There are several methods to prepare GO/metal oxide composites with the same starting step of GO synthesis. After GO synthesis, Fe₃O₄ (pre-prepared by coprecipitation and functionalized with -NH₂ groups of silane compounds) is encapsulated to the GO surface with -COOH groups activated by EDC and NHS by CO-NH covalent bonds (He *et al.*, 2010; Xie *et al.*, 2012). The Fe₃O₄/GO composite can be synthesized by thermal decomposition of iron (III) acetate in ethylene glycol solvent in the presence of GO (Farghali *et al.*, 2015). Another way to incorporate Fe₃O₄ into GO is the coprecipitation of an Fe(III) and Fe(II) acidic solution consisting of GO by 1M NaOH solution (Zubir *et al.*, 2014). Fe₃O₄ and GO can be synthesized separately and then combined (at a mass ratio of 1:1) into one composite material through calcination in a Teflon-lined autoclave for 8 h at 200°C. The magnetic GO can then be further modified with EDTA by mechanical rubbing at room temperature for 24 h (Cui *et al.*, 2015). A GO-Fe₂O₃ hybrid material was obtained by aging the suspension of GO containing Fe(NO₃)₃ for 8 h at 60°C (Guo *et al.*, 2013). Some authors also combined both metal oxide and a polymeric matrix to GO materials by coprecipitation of Fe₃O₄ in a suspension of GO in the presence of chitosan to form a CS/Fe₃O₄/GO composite (Tran *et al.*, 2017). To obtain a GO-MnFe₂O₄

magnetic composite, ferric chloride ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$) and manganese sulfate ($\text{MnSO}_4 \cdot \text{H}_2\text{O}$) were added to the colloidal GO solution and continuously stirred for 30-60 minutes at the temperature of 80°C and pH of 10.0-10.5 adjusted by NaOH or ammonia solution. The obtained precipitate was magnetically isolated, washed with water and methanol, and finally dried overnight (Siddiqui & Chaudhry, 2018).

Another metal oxide commonly fused with GO is TiO_2 . The GO- TiO_2 composite can be synthesized by the liquid phase deposition (LPD) method. In short, ammonium hexafluorotitanate (IV) and boric acid are added to a certain amount of the GO dispersion at 60°C for 2 h under vigorous stirring. The composite is obtained after filtration, washing with water, and vacuum drying at 100°C (Cruz *et al.*, 2015). GO- TiO_2 can also be added to the membranes of various polymers such as an anionic perfluorinated polymer, cellulose acetate, polycarbonate, polysulfone fluoride (PSF), or polyvinylidene fluoride (PVDF). The polymeric membranes containing GO- TiO_2 have outstanding chemical resistance, thermal stability, good membrane formation, and excellent photocatalytic effects (Dadvar *et al.*, 2017).

3. WATER TREATMENT APPLICATIONS

Graphene oxide can be used for an abundant of environmental applications, i.e. adsorption (for heavy metal ions, organic compounds, gases), as photocatalysts (for metal ion reduction, dye degradation or microorganism), as desalination membranes, or as electrodes for environmental sensors (Mauter & Elimelech, 2008; Gadipelli & Guo, 2015; Li *et al.*, 2015). In this review, only important applications for contaminated water are discussed.

3.1. Adsorption

In general, the adsorption equilibrium is often described by mathematical models. Among which, three classic adsorption models, namely the Langmuir, Freundlich, and Temkin models,

are the most frequently used. The Langmuir (1), Freundlich (2) and Temkin (3) equations are shown below:

$$\frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{C_e}{q_m} \quad (1)$$

$$\ln q_e = \frac{1}{n} \ln C_e + \ln K_F q_m \quad (2)$$

$$q_e = K_T \ln C_e + K_T \ln I_f \quad (3)$$

where q_m is the theoretical maximum adsorption capacity per unit weight of the adsorbent (mg g^{-1}), K_L , K_F , and K_T are the adsorption constants of the Langmuir, Freundlich and Temkin models, respectively, and n is the Freundlich linearity index (Dada *et al.*, 2012).

3.1.1. Heavy metal ions

A polypyrrole-GO nanocomposite with the zero charge pH of 6.2 can be used to adsorb Cr(VI) ions in an aqueous medium both in batch and packed-bed column modes. The Langmuir maximum capacity of 625 mg g^{-1} at 25°C was found. Other ions in the solutions did not affect Cr(VI) adsorption. Especially, the column mode experiments showed that 2 g of the nanocomposite can remove 10 mg L^{-1} Cr(VI) in a large volume (64.08 L) at a flow rate of 3 mL min^{-1} (Zebedius *et al.*, 2015).

Schwertmannite, a mineral of Fe (III)-oxyhydroxy sulfate with different compositions, usually expressed as $\text{Fe}_8\text{O}_8(\text{OH})_{8-2x}(\text{SO}_4)_x$ (in which x is within 1 to 1.75), can be incorporated with GO to form GO-SCH composites which can be used to adsorb Sb(V). Several water sources containing Sb(V) up to $50,000 \text{ } \mu\text{g L}^{-1}$ were treated with GO-SCH and the Sb(VI) in all the samples was removed to below regulation levels for these waters (below $5 \text{ } \mu\text{g L}^{-1}$, $100 \text{ } \mu\text{g L}^{-1}$, and $1,000 \text{ } \mu\text{g L}^{-1}$ for spiked tap water, simulated river water, and acid mine drainage, respectively) (Dong *et al.*, 2015).

Arsenic can be removed (over 99.9% within 1ppb) from water by magnetite-reduced graphene oxide nanocomposites (m-rGO). The composites consist of 10 nm Fe_3O_4 particles that allow isolation of the material from an aqueous

medium by a magnet. Because of more active sites on the surface of reduced graphene oxide, the m-rGO demonstrate such a high adsorption capacity for both As(III) and As(V) that m-rGO could be used for treating drinking groundwater in many Asian countries (Chandra *et al.*, 2010).

Magnetic-GO (mGO) is also widely modified for use in heavy metal adsorption. EDTA-mGO performs at the high efficiencies of 96.2%, 95.1%, and 96.5% for Pb(II), Hg(II), and Cu(II) removal, correspondingly. The adsorption data are well described by the pseudo-second-order model and fitted to the Freundlich and Temkin models. From the Langmuir isotherm model, the maximum adsorption capacity is 508.4 mg g⁻¹ for Pb(II), 268.4 mg g⁻¹ for Hg(II), and 301.2 mg g⁻¹ for Cu(II). Thermodynamic data were calculated ($\Delta G < 0$, $\Delta H > 0$, $\Delta S > 0$) and suggest that the adsorption process is endothermic and spontaneous. Moreover, the adsorption ability of EDTA-mGO changed insignificantly through 5 cycles of the experiment (Cui *et al.*, 2015). Another GO/Fe₃O₄ nanocomposite material synthesized by slurry mixing had a large specific surface area (72.9 m² g⁻¹) and the adsorption capacity for Pb²⁺ ions (54.64 mg g⁻¹) (Hieu *et al.*, 2015).

3.1.2. Organic dyes

The authors of (Anh *et al.*, 2015) used graphene oxide to adsorb RR195 dye and found that the adsorption capacity of graphene oxide is 212.7 mg g⁻¹ and the adsorption obeys the second order Langmuir isothermal model.

GO-CS was reported to be among the best adsorbents of MB with an adsorption capacity of 4.68x10² mg g⁻¹ and the ability to decolorize the 100 mg L⁻¹ MB solution to colorless. The adsorption process was well fitted to the Freundlich model. The materials after adsorption could easily be removed by low-speed centrifugation (Yang *et al.*, 2013).

When used as dye adsorbents, mGO materials usually contain a high Fe₃O₄ content that helps to easily separate the materials from the environment. Containing 23.6% Fe₃O₄, a GO/Fe₃O₄ composite is capable of adsorbing

167.2 mg g⁻¹ of MB after 30 minutes and 171.3 mg g⁻¹ of Neutral Red (NR) after 90 minutes (Xie *et al.*, 2012). As the Fe₃O₄ content increases, the GO/Fe₃O₄ adsorption capacity decreases (Farghali *et al.*, 2015). Containing 54% Fe₃O₄ and 10% GO by mass, a CS/Fe₃O₄/GO nanocomposite with an average size of Fe₃O₄ particles of 30-40 nm adsorb MB obeying the Langmuir isotherm model with the adsorption capacity of only 30.1 mg g⁻¹ (Tran *et al.*, 2017).

There are also studies in which mGO was used to adsorb a heavy metal ion (Cd(II)) and organic dyes (MB and OG) simultaneously. The results showed that the maximum sorption rates of mGO for single Cd(II), MB, and OG solutions were 91.29 mg g⁻¹, 64.23 mg g⁻¹ and 20.85 mg g⁻¹, respectively. In the 2-component model, the sorption capacity increases for OG but changes little for MB when the Cd(II) concentration is increased. The sorption capacity for Cd(II) decreases when more MB is added but does not depend on the concentration of OG (Deng *et al.*, 2013).

Some parameters of heavy metal and organic dye adsorption are summarized in table 1.

3.1.3. Other organic compounds (antibiotics, aromatic hydrocarbons)

Besides heavy metal ions and organic dyes, other organic compounds like antibiotics and aromatic compounds can be removed from a water medium by GO and GO-based materials.

A suspension of GO was introduced to certain concentration solutions of tetracycline. The results showed that the adsorption of tetracycline on the surface of the GO was obtained through π - π interactions between rings in the tetracycline molecule and the hexagonal cells of the graphene oxide and cation - π bonding. The isothermal adsorption obeyed the Langmuir and Temkin models with the maximum adsorption capacity of $q_m = 313$ mg g⁻¹ that confirms GO is an excellent adsorbent. The adsorption performed better at low pH and Na⁺ concentrations. In a comparative

experiment with other antibiotics, it was found that $q_m \text{ doxycycline} > q_m \text{ oxytetracycline} \approx q_m \text{ tetracycline}$ due to the different pKa values of the antibiotics (Gao *et al.*, 2012). Another antibiotic, ciprofloxacin, was also investigated for its removal from an aquatic environment by GO/SA hydrogels and aerogels. The adsorption capacities of the GO/SA gels were improved by their porous properties, π - π bonding, and C = O groups on the surface of GO (Fei *et al.*, 2016).

Five polycyclic aromatic hydrocarbons (fluoranthene, pyrene, benzo[a]anthracene, benzo[b]fluoranthene, and benzo[a]pyrene) were shown to be effectively extracted from a water environment by a $\text{Fe}_3\text{O}_4/\text{GO}$ nanocomposite as a magnetic solid-phase extraction sorbent. The excellent adsorption properties of the $\text{Fe}_3\text{O}_4/\text{GO}$ nanocomposite resulted from the principal roles of π - π stacking interactions and hydrophobic interactions between the hydrocarbons and the graphene sheet. It was observed that the recoveries of the hydrocarbons from the $\text{Fe}_3\text{O}_4/\text{GO}$ sorbent ranged from 76.8% to 101.2% (Han *et al.*, 2012).

3.2. Catalyst

3.2.1. Photocatalyst

TiO_2 is a well-known photocatalyst for the degradation of a wide range of pollutants. Incorporating TiO_2 with GO may lead to the much better effectiveness of TiO_2 catalysts. Comparative experiments of photodegradation of a 4-pesticide mixture (diuron, alachlor, isoproturon, and atrazine) in the presence of TiO_2 and GO- TiO_2 were carried out. The results revealed that TiO_2 catalytic activity in natural water was much lower than that in pure water. On the other hand, GO- TiO_2 exhibited high activity in both water matrices. In addition, GO- TiO_2 provided to have higher photocatalytic activity than TiO_2 under visible light (Cruz *et al.*, 2015).

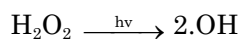
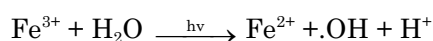
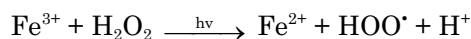
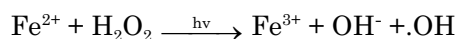
Other authors prepared polymeric membranes consisting of GO- TiO_2 to enhance the filtration efficiency of the membranes. In addition, the photocatalytic property of TiO_2 allowed organic and odorous compounds in

water to be degraded. Therefore, the cost of washing membranes could be reduced significantly with the produced membranes. The energy used by the membranes also decreased by half compared to the standard nanomembranes (Dadvar *et al.*, 2017).

Under visible light, GO/graphitic- C_3N_4 exhibited efficient photocatalytic capability to degrade rhodamine B and 2, 4-dichlorophenol. GO was reported to act as a separation center and electron acceptor to increase the catalytic activity of GO/g- C_3N_4 from 2 to 3.8 times compared to pristine g- C_3N_4 (Liao *et al.*, 2012).

3.2.2. Advanced oxidation degradation: Photo-Fenton and Fenton-like processes

Photo-Fenton and Fenton-like processes are reactions related to the catalytic activity of iron ions and are usually described by the following mechanism (Gonzalez-olmos *et al.*, 2012):



$\cdot\text{OH} + \text{organic compound} \rightarrow \text{oxidized products}$

For this purpose, Fe_3O_4 -GO nanocomposites with different GO compositions (from 0 to 15%) were synthesized and characterized. The Fe_3O_4 -GO was assessed for photo-Fenton degradation of phenol in different experimental conditions, such as catalyst dosage, H_2O_2 initial concentration, and initial pH of the phenol solution. The authors found that the optimal conditions (pH 5.0, H_2O_2 concentration 10.0 mmol L^{-1} , and catalyst dosage 0.25 g L^{-1}) combined with UV-light irradiation resulted in around 98.8% phenol and 81.3% TOC removal after 120 mins. It was also reported that Fe_3O_4 -GO still performed high catalytic activity after five cycles. Especially, GO was stated to improve the catalytic activity of Fe_3O_4 by accelerating the adsorption capacity, providing more active sites, and increasing the UV light irradiation (Yu *et al.*, 2016).

Table 1. Adsorption parameters of graphene oxide based materials

Adsorbent	Contaminant	Contaminant initial conc. range (mg L ⁻¹)	pH (optimal pH)	Mass of adsorbent/ volume of contaminant solution	Model	Main result q _m (mg g ⁻¹)	Desorption substances	References
GO	Co(II)	2-25	2-8 (5.5)	1.0 g L ⁻¹	Freundlich	21.28		Prasanna <i>et al.</i> , 2015
Polypyrrole-GO	Cr (VI)	100-500	2-9 (2)	0.025g/50 mL	Langmuir	625		Zebedius <i>et al.</i> 2015
GO-SCH	Sb(VI)	0-55	3-10.5 (7)	0.3 g L ⁻¹	Langmuir	158.6	HCl 0.1M EDTA 0.1 M-5 cycles	Dong <i>et al.</i> , 2015
m-rGO	As(III) As(V)	3-7	4-10 (4)	0.2 g L ⁻¹	Langmuir Freundlich	~ 10 ~ 5		Chandra <i>et al.</i> , 2010
EDTA-mGO	Pb(II) Hg(II) Cu(II)	100	1-7	2-16 mg/25 mL	Freundlich Temkin	508.4 268.4 301.2	HCl, NaOH or Ca(NO ₃) ₂ (0.1 M)-5 cycles	Cui <i>et al.</i> , 2015
PET/PAN/ GO/Fe ₃ O ₄	Pb(II) Cr(VI)	20-1000	2-7 (6) 2-8 (3)	0.5 g L ⁻¹	Redlich-Peterson	799.4 911.9	HCl 1M 5 cycles	Koushkbaghi <i>et al.</i> , 2016
GO/Activated carbon felt	U (VI)	0-50	1-10 (5.5)	0.01 g/50 mL	Langmuir	298		Chen <i>et al.</i> , 2013
GO	MB	400-4000	3-11	10 mg/25 mL	Langmuir	1.939 mg/mg	CH ₃ COOH/NH ₃ in methanol (30-40%)	Zhang <i>et al.</i> , 2011
Chitin/GO	Remazol black (RB) Neutral red (NR)	0.025-5 mM 0.025-7 mM	4-8 (5) 4-8 (4)	1 mg mL ⁻¹	Sips	0.093 mmol/g 0.057 mmol/g	Buffer solution pH 7-10 60% (BB), 100% (NR)	González <i>et al.</i> , 2015
A-rGO/ Co ₃ O ₄	Rhodamine B (RhB)	5-200	2-12 (2)	0.01 g/10 mL	Freundlich	102.9		Altaa <i>et al.</i> , 2018
Graphene/ Fe ₃ O ₄	MB	10-25	2-11	0.01 g/25 mL	Langmuir	43.82	Ethanol 5 cycles	Ai <i>et al.</i> , 2011
PS/Fe ₃ O ₄ / GO	RhB	0-150		2 mg mL ⁻¹		13.8		Wang <i>et al.</i> , 2012

Another report showed that with a GO/Fe₃O₄ Fenton-like catalyst, Acid Orange 7 (AO7) was decomposed effectively. A series of GO/Fe₃O₄ composites with a GO composition range from 5 to 25% were investigated and the results showed that with a 10% GO composition, the materials achieved the highest BET surface area, while the 5% GO sample was the best catalyst that was initially equal to and then higher than bare Fe₃O₄ (Zubir *et al.*, 2014). The reaction conditions using the material as a Fenton-like catalyst were also optimized to be 0.2 g/l at a pH solution of 3, room temperature, and H₂O₂ concentration of 22 mM. The mechanism of the reaction could be well fitted to the Langmuir-Hinshelwood pseudo-first-order kinetic (Zubir *et al.*, 2014). In addition, the GO/Fe₃O₄ catalyst can be recycled (up to 7 times in use) thanks to the electron donor-acceptor properties of GO that prevented the active sites (Fe²⁺) of Fe₃O₄ from being oxidized in the heterogeneous Fenton-like reaction (Aida Zubir *et al.*, 2015). Besides Fe₃O₄, the GO-based nanocomposite system was also synthesized with CoFe₂O₄ for Fenton heterogeneous and photo Fenton catalytic processing (Hoa *et al.*, 2015). Similarly, ZnFe₂O₄-rGO was used for the catalytic removal of MB. Two hours after adding 10 mg of this material to 50 mL of a 10 mg L⁻¹ MB solution under a 150 W halogen lamp light, about 80% of the MB was removed. The catalyst could be reused for 5 cycles (Jumeri *et al.*, 2014).

The photocatalytic activities of GO-Fe₂O₃ hybrid materials were evaluated by the degradation of Rhodamine B (RhB) in an aqueous medium under visible irradiation (>420 nm). It was found that the catalyst was effectively able to decompose RhB at various conditions of solution pH (from 2.09 to 10.09). The catalyst dosage and H₂O₂ concentration for optimal degradation were also determined. Notably, it maintained high activity after 7 cycles. GO-Fe₂O₃ was also efficient in degrading 4-nitrophenol (4-NP). The catalytic mechanism was described with the main degradation occurring on the surface of the catalyst without

significant iron leaching and involvement of the hydroxyl radicals during the process (Guo *et al.*, 2013). In addition, 4-NP was reduced by NaBH₄ with the catalyst of another hybrid material, rGO/Fe₃O₄/Ag. The catalytic activity retained 94.4% after the fourth cycle (Thu, 2017).

3.3. Antimicrobial effects

GO is considered to be a novel alternative for antibiotics to combat multidrug-resistance bacteria (Yousefi *et al.*, 2016). The antibacterial activity of GO against the bacterial model of *Escherichia coli* was investigated in comparison with graphite (Gt), graphite oxide (GtO), and rGO. The results showed that the highest antibacterial activity belonged to GO and was sequentially followed by rGO, Gt, and GtO. The morphology determined by scanning electronic microscopy revealed that most of the *E. coli* cells treated with GO become flattened with irreversibly damaged cell membranes, leading to the destruction of cell structures (Liu *et al.*, 2011). The mechanism was described as GO causing the outer and inner cell membranes of *E. coli* and *E. faecalis* bacteria to be destroyed leading to the release of adenine and protein from the bacteria (Nanda *et al.*, 2016). It was also reported that the antimicrobial activity of GO sheets is deeply dependent on their size. Smaller GO sheets may have higher activity thanks to the oxidative mechanisms involved in the higher defect density of smaller sheets (Perreault *et al.*, 2015).

The synergetic antimicrobial effects of GO and Ag nanoparticles in GO-Ag nanocomposites have attracted much attention from researchers. GO-Ag nanocomposites express much higher activity against both Gram-negative and Gram-positive bacterial strains compared to AgNPs. The increase in antibacterial activity of GO-AgNPs is probably due to the high stability of AgNPs attached to GO sheets and the positively charged surface of the composite that enhances their electrostatic interaction with the negatively charged bacterial cell membrane (Zhu *et al.*, 2013). Various contents of Ag were examined in GO-

Ag composites for their best bioactivity. It was found that at 0.2 M AgNO₃, GO-Ag obtained a 48.77% antimicrobial activity that was higher than other concentrations (Vi & Lue, 2016). Low pH, high dosage, and prolonged treatment time favor the disinfection rate of GO-Ag composites. AgNPs presented on the GO sheets were first oxidized into silver ions which then killed the bacteria. Superoxide anion radicals and hydrogen peroxide then formed which also contributed to the antibacterial activity (Song *et al.*, 2016). In addition, GO-TiO₂-Ag nanocomposites can not only effectively disinfect but can also decontaminate under solar irradiation (Liu *et al.*, 2013). Details of several disinfection processes of GO-based materials are listed in Table 2.

4. CONCLUSIONS

In conclusion, the number of studies on GO and GO-based materials has increased significantly in recent years. After the initial step of GO synthesis, various GO-polymers and GO-

metal/metal oxide materials have been prepared by different methods. Thanks to the valuable properties of GO and expected properties of the other components, the GO-based materials can be widely applied in environmental treatments. They are excellent adsorbents for heavy metal ions and a number of organic compounds. They might be considered as an outstanding catalysts for photodegradation or Fenton/Fenton-like reactions. Moreover, some GO-based materials exhibit significant antibacterial activities. These facts confirm the potential of GO-based materials for water purification. However, to optimize the performance of GO and its derivatives, and enlarge their applications, it is essential for further investigations to be conducted.

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Table 2. Disinfection details of several GO-based materials

GO-based material	Bacteria	% survival	Main mechanism	References
GO 100-400 µg mL ⁻¹	<i>E. coli</i> K-12, <i>L.acidophilus</i> ADH, <i>B.animalis</i> Bif-6 Caco-2	Biocompatible	GO agglomeration	(Nguyen <i>et al.</i> , 2015)
GO 0.25 mg mL ⁻¹	<i>Klebsiella pneumoniae</i> <i>E. coli</i> <i>P. aeruginos</i>	2.8 11.8 11.9		(Wu <i>et al.</i> , 2017)
GO and graphene nanowalls	<i>E. coli</i> <i>Staphylococcus aureus</i>	16-41 5-26	cell membrane damage	(Akhavan & Ghaderi, 2010)
GO-hydroxyapatite-TiO ₂	<i>E. coli</i> <i>Staphylococcus aureus</i>	37-98 26-86	RNA leakage, irreversible membrane damage	(Parcharoen <i>et al.</i> 2017)
GO/Ag (100 ppm GO, 100 ppm Ag)	<i>E. coli</i> <i>Staphylococcus aureus</i>	0 0	Membrane stress, oxidative stress	(Tam <i>et al.</i> , 2016)
rGO/Ag 100 µg mL ⁻¹	<i>Proteus mirabilis</i> <i>E. coli</i> <i>Staphylococcus aureus</i>	23 mm 25 mm 24 mm*	Synergic effect	(Prasad <i>et al.</i> , 2017)
PVK-GO 30 µg mL ⁻¹	<i>Escherichia coli</i> <i>Cupriavidus etalliduran</i> <i>Bacillus subtilis</i> <i>Rhodococcus opacus</i>	0 0 0 0	interrupting the metabolic activity	(Mejías <i>et al.</i> , 2012)

Note: * average zone of inhibition

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