A brief review on recent graphene oxide-based material nanocomposites: Synthesis and applications

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Abstract

In a past few years, more focus has been given to graphene, especially for its facile synthesis, novel hybrids materials and applications. Recently, researchers' attention has also focused on graphene oxide (GO) and reduced graphene oxide (rGO) nanocomposites, which lead to the development of various applications. Their superb and impressive characteristic makes them suitable candidates to be hybridized with polymer, metal oxide, and biomaterials. In this brief review, we will run through the accomplishments of effective approaches for synthesizing graphene oxide-based material nanocomposites together with their recently developed applications.

Keywords: Graphene oxide, Reduce graphene oxide, Synthesis, Application

Table of Content

1. Introduction

Graphene oxide (GO) contains a cluster of reactive oxygen functional groups, which make it a strong nominee for use in many applications through chemical functionalization. GO is constructed from graphite oxide and it has been a favorable passage for producing a large scale production of graphene [1–3]. Graphene, however, has a major drawbacklow dispersibility in water, causing its surface area to decrease, and therefore, limits its application. This is due to aggregation that is caused by the strong van der Waals interactions and π–π stacking of the graphene sheets [4].

Therefore, interest has been concentrated on assimilating GO with other materials by hybridizing it with good water-dispersibility materials [5]. By introducing functional groups on the graphene sheets via chemical oxidation, aggregation can be cut down or eradicated, which leads to fabrication of graphene oxide [6]. The dispersibility of GO only increases in polar solvent due to the hydrophilic functional groups [7]. In addition, controlled oxidation provides tunability of the electronic and mechanical properties, including the possibility of accessing zero-band gap graphene via a complete removal of the C-O bonds [8]. Various functionalities on the surface of GO make it an ideal platform for chemical modification, which may generate materials with amazing properties.

Aside from the advanced properties, the detailed chemical structure of GO has been the focus of a fascinating debate over the years, and yet, still no confirmed model has been proposed. The only common information is the structural characteristics of GO, which is the presence of many oxygen functional groups on the GO's surface such as epoxy, hydroxyl, and the carboxylic acid group [9]. Due to the deficiency of precise analytical techniques for characterizing the amorphous property of GO, and nonstoichiometric berthollide character of GO, searching for a real model of GO becomes problematic [1].

A structural model has already been proposed by many researchers such as Hofmann and Holst, Ruess, Scholz-Boehm, and Nakajima-Matsuo [1]. Based on all of these models, GO is generally constructed by regular lattice, consisting of a discrete repeated unit. Moreover, the atomic and electronic structure of GO is elucidated by Mkhoyan et al. [8] by measuring the structure of C and O K-edges. It is reported that the GO sheet shows a surface roughness about 0.6 nm and the structure is mainly amorphous due to distortion from sp^3 C-O bonds. The results also reported that a ratio of 1:5 oxygen to carbon atoms is sufficient to transform the measured 40% of the carbon bonds into sp^3 bonds [8]. Erickson et al. [10] shared almost the same opinion and adding up a few more detail of the structure of GO. They found that the graphitic region is up to 8 nm^2 and a hole of about 5 nm is present on the GO sheet. Besides that, the unstrained sp^2 bonds present between carbon, forming a continuous network across the GO sheet and the oxidized region, exhibit no order [10].

The synthesizing method of graphene oxide was already developed in 1859 [1]. At that time, Brodie performed a reaction between graphite and $KClO_3$ in fuming HNO_3, which resulted in a material with an increase in the mass of flake graphite. Later, in 1898 Staudenmaier further the previous method by raising the acidity of the mixture using concentrated H_2SO_4 and adding the chlorate in several aliquots over the course of the reaction [1]. After that, Hummer and Offeman improved the method using alternate oxidation. The reaction involved the oxidation of graphite by using $NaNO_3$, KMnO4 and concentrated H_2SO [1,11]. This method is mostly used now with some modifications and improvements.

Marcano et al. [12] developed a synthesizing method that improves the efficiency of the oxidation process by excluding the presence of $NaNO_3$, raising the usage of $KMnO_4$, and using the ratio 9:1 mixture of H_2SO_4/H_3PO_4. This method was confirmed to generate a larger amount of hydrophilic GO material compared to the conventional Hummer method; also, it does not release toxic gas and easily controls the temperature [12]. Yang et al. [13] also took some modifications of Hummer's method to develop a facile synthesis method of GO by using expanded graphite oxide as a starting material. This method intensely decreases acid, is extremely time

saving, consumes minimal energy, and has high efficiency without releasing toxic gas [13]. Until now, continuous, rapid research has evolved to develop simple and greener methods to prepare GO.

2. Synthesis of Graphene Oxide-based Material Nanocomposite

There is great attention to the preparation method of GO-based material nanocomposites. GO-based material nanocomposites can be synthesized by different methods and approaches including the hydrothermal method [14,15], electrochemical codeposition [16], in situ polymerization [17,18], microwave-assisted method [19,20], vacuum impregnation [21], and sol gel technique [22]. In the GO-based nanocomposite, GO presents either as a functional component or as a substrate for immobilizing the other components [23,24]. Therefore, this section will mainly focus on an effective synthesis method that has been practiced by some researchers.

2.1. Hydrothermal Method

The hydrothermal method is an effective and frequently favored method to synthesize GO-based nanomaterial, which is performed in high temperature and vapor pressure. This method is a very low cost method that is facile, environmental friendly, and an applicable route to synthesize soluble reduce graphene oxide (rGO) sheets in excessive amounts [25]. This method has been used by Xu et al. [14] in a one-pot approach to synthesize $rGO/CoWO_4$ and results in an enhanced electrochemical performance for supercapacitor. Similarly, Dong et al. [26] also use the same method to synthesize rGO/TiO_2 nanocomposite as anode material for lithium ion batteries. Hydrothermal, also known as the solvothermal method, is usually carried out in a Teflon-line autoclave and the temperature is between 160 and 180 °C[14,27,28].

Later, this method was modified, and microwave-assisted hydrothermal method was developed. Gui et al. [29] successfully used the microwave-assisted hydrothermal method to synthesize graphene/WO_3 by employing GO as a starting material. This method was upheld as a greener approach that involves a lower temperature and minimizes the duration of the reaction compared to the conventional hydrothermal method [29]. This method as well has been manipulated by Tang et al. [30] to prepare rGO/SnO_2 nanocomposites, assisted by an ionic liquid that acts as a reaction medium in order to enhance the microwave-assisted hydrothermal method. The ionic liquid, 1-butyl-3-methylimidazolium tetrafluoroborate ([bmim]BF4) that is used in this reaction is a greener solvent compared to the established organic solvent in addition to being supported by the excellent solvent power, lower vapor pressure, its being non-flammable, having good thermal stability, its ionic conductivity, and high electrochemical stability [30].

Another option of hydrothermal method is by using a continuous hydrothermal flow synthesis (CHFS) reactor. It can manipulate other reaction variables like pressure and temperature and also particle properties involving mixing a flow of supercritical water with a flow of aqueous metal salt, thus resulting in a rapid precipitation and controlled growth of nanoparticles [31]. This method is simply one of the most economical approaches that can control the size of the product by enhancing the heating rate, as well as saving time [30].

2.2. Electrochemical Deposition

Electrochemical deposition is a process by which a material is deposited from a solution of ions onto the surface of the electrical conductor or electrode. Primarily, this method is employed to fabricate the electrochemical sensor by depositing nanocomposite material onto an electrode [24,32,33]. Du et al. [32] has employed the one-step electrodeposition method to synthesize a novel glucose sensor based on the rGO-based nanocomposite. In this study, dendritic gold nanostructure is hybridized with rGO functionalized with a globular protein, β-lactoglobulin, and was electrodeposited on a glassy carbon electrode (GCE) by the chronoamperometry technique [32]. Similarly, Wu et al. [34] modified Au electrode by direct electrodeposition on rGO and followed by the chronoamperometry technique; then, electrochemical polymerization takes place to modify the electrode with poly(3-aminophenylboronic acid) film.

Reza et al. [33] also use this method to deposit rGO/Cn nanocomposites on an indium tin oxide (ITO) glass substrate. The preparation of electrodeposited electrodes involves two electrodes that were immersed in colloidal rGO/Cn nanocomposite suspension where platinum foil acts as a cathode and ITO glass substrate acts as an anode [33]. Apart from the electrochemical electrode and sensor, this method also can assist the preparation of coated film. This study has been done by Li et al. [16], which has synthesized GO/ZnO nanocomposite films on Fluorine doped Tin Oxide (FTO) coated glass, resulting in improved photoelectric conversion properties as a photoabsorber.

2.3. In Situ Polymerization

In situ polymerization is a prevalent route for the preparation of GO-based polymer nanocomposite on a larger scale and avoiding the chain destruction [35]. It has been used to prepare polyaniline, polypyrrole, and polymethyl-methacrylate in situ on a GO and rGO layer [17,18,35–38]. Taking polyaniline as an example, the distinctive precursor used is aniline, which is then immersed in HCl solution with the addition of ammonium persulfate that acts as an oxidant [17,36,37].

Similarly, Yan et al. [35] prepared polypyrrole/SDBS (sodium dodecyl benzenesulfonate) intercalated reduced graphene oxide (rGO) nanocomposites by using pyrrole as a monomer. In the first step, SDBS is intercalated with GO and it is then reduced by hydrazine at 95 °C and ammonium persulfate is added to initiate the polymerization of pyrrole monomer. There are intermolecular forces involving π-π stacking, hydrogen bond and van der Waals forces that promote the adsorption of pyrrole monomer onto the rGO surface. These intermolecular forces between aromatic rings of polypyrrole chains and sp^2-bonded carbon atoms of graphene basal planes also make the polypyrrole coat perfectly onto the sheets of graphene [35]. The benefit of an in situ polymerization method lies in the fact that the whole procedure is facile, controllable, and scalable, promoting the intercalation of monomer between the GO and rGO sheets.

2.4. Sol-gel Technique

Recently, the sol-gel technique has been used to synthesize GO-based material nanocomposite especially in the fabrication of self-cleaning film and glass coating. Being the fact that sol-gel derived inorganic composites require a low synthesis temperature, the physical properties of the doping component could be retained by controlling the mixture between the guest molecule and the host matrices [39].

Azarang et al. [40,41] has synthesized a photocatalyst based on rGO/ZnO nanocomposite by using sol-gel technique. It is reported that gelatin medium (from bovine skin) is used as a polymerization agent along with GO, and $Zn(NO_3)_2.6H_2O$, as a starting materials. In the first step, GO was added into the zinc nitrate solution followed by the addition of gelatin solution. The solution was stirred at 80 °C until a dark brown gel was formed. Then the gel was calcined and followed by post-anneling process under Ar gas [40]. The same procedure was adopted by He [22] with the utilization of ethylene glycol as a polymerization agent. Then, the mixed solutions were dip-coated on the ultrasonically glass substrate to produce a self-cleaning glass. Later, the same method was also reported by Azarang et al. [42] using starch as the polymerization agent. The growth of the ZnO nanoparticle on rGO is terminated by long-chain starch compounds. The annealing process of the resulting products is carried out at 350 °C to remove the starch and produce a rGO sheet in one-pot without any post-annealing processes.

The sol-gel approach offers a distinctive means to prepare a three-dimensional network capabilityfor the encapsulation of numerous biomolecules. Peng, Huang, and Zheng [43] have synthesized ferrocene-branched organically modified silica material ormosil/chitosan/GO nanocomposite for a glucose sensor. Ormosil is contributed in modification of surface property of the matrices. It enhances the film flexibility and prevents leakage when the reagent is attached to the silica [43]. Furthermore, sol-gel derived silica glasses also show an outstanding optical, thermal, transparency in the UV region, and high thresholds for laser damage [39].

2.5. Sonochemical by Ultrasonic

Reducing GO and rGO under a mild condition become a great challenge especially when no chemical reduction and/or stabilizing agents is involve. Therefore, the chemical upshot of ultrasound irradiation treatment evolve from the acoustic cavitation occurrence [44]. The sonochemical method offers a severe condition for reducing GO and metal ion precursor that normally carried out at 20 kHz using a titanium horn, where the shear forces generated by the acoustic cavitation are enough to overcome the van der Waals forces between the graphene sheets and prevent their reaggregation [45].

The mechanism of sonochemical treatment initiate from the irradiation of the liquid with ultrasound. The bubbles are generated and pile up the ultrasonic energy with their growth and subsequent collapse which generate the accumulated energy within a split second. These cavitational implosions generate localized hotspots with a high temperature up to 5000 K, pressure of 1000 bar, and heating and cooling rate of 1010 K/s [44,46,47]. These are the promising conditions lead to the formation of smaller sized inorganic particles and helpful to reducing the agglomeration of nanoparticles and the restacks of the exfoliated graphene sheets [48].

Anandan, Manivel and Ashokkumar[49]successfully prepared Pt supported Sn on rGO sheets through a sonochemical approach. The nanoparticle precursors dispersed on the graphene sheets were prepared by the

simultaneous sonochemical reduction procedure with low frequency ultrasound at 20 kHz. Recently, Golsheikh et al. [44] applied the same method to decorate a uniform hierarchical ZnS nanospheres onto the surface of rGO. Similarly, Peng et al.[48] synthesized ZnO/rGO photocatalyst through sonochemical method which resulting an increasing photocatalytic activity of ZnO and prevent the photocorrosion. Ultrasound assisted technique also leads to a good control on the ultimate morphologies of ZnO nanoparticles [47]. This method also has assist the development of DNA biosensor through the preparation of toluene blue/GO nanocomposite[50].Dezfuli et al. [51] also adopted the same approach in synthesizing the CeO_2/rGO nanocomposites. The results show that the uniformly size distribution of CeO_2 nanoparticles well anchored on rGO. It reveals that the CeO_2/rGO nanocomposites exhibit a synergistic effect at a suitable loading content of CeO_2 on rGO.

2.6. Electrospinning

Electrospinning has been widely used as a versatile technique for the preparation of nanocomposite nanofibers with a diameter ranging from several micrometers down to tens of nanometers [52,53]. It is an effective technique for making nanofibers due to its simplicity and flexibility in producing homogeneous fibers with an adjustable diameter and a microstructure with various nanofillers [54].

In the recent past, various nanocomposites have been fabricated by the electrospinning approach, for instance, GO/polyaniline/polyvinylidene fluoride nanofibers [55], GO/poly(vinyl alcohol) [54], GO/poly(vinyl alcohol)/TiO_2 [56], polyacrylonitrile/GO [57], and GO/vanadium pentoxide [58]. In general GO composite suspensions were filled in a 5 mL syringe with a blunt-end, stainless steel needle attached at the open end. Then, a high DC voltage in the range of 0–50 kV was supplied to the needle. An aluminum foil was used as the collection screen and it was connected to the ground electrode of the power supply. The electrospinning process was carried out at room temperature. The obtained nanofibers were post-annealed at 500 °C under N_2 and H_2 to reduce the GO in the nanofibers [54].

The polyacrylonitrile/GO composite nanofibers were also successfully fabricated using electrospinning approach [57]. The procedure is very similar to the one used by Wang et al. [54] where the composite electrospinning solution was electrospun at a positive voltage of 15 kV with a working distance of 15 cm, and the flow rate was set as 1.5 mL/h. Based on transmission electron microscopy (TEM) the clusters of GO nanoplatelets are clearly well incorporated into the polymer matrix and oriented in the fiber axial direction. This is due to the higher draw ratio that imparted a larger stress on the fiber as it was being formed during the electrospinning process, and gave a proper alignment of the two-dimensional GO pallets along the fiber axis nanostructures and surface nanomechanical properties of polyacrylonitrile/graphene oxide composite [57]. The same morphology is obtained by GO/vanadium pentoxide composite nanofibers that have smooth and uniform surfaces [58].

2.7. Microwave

Microwave synthesis has lately been shown to have a big impact, is more environmental friendly, and less energy is required compared to conventional heating for the synthesis of inorganic nanomaterials by soft chemistry [19,59]. This technique offers a more homogeneous heating process and can speed up the reaction rate by orders of magnitude [60]. It can heat the reactant to a high temperature very quickly by transferring energy selectively to microwave absorbing polar solvents with a simultaneous increase in self-generated pressure inside the sealed reaction vessel [61]. As is widely known, microwave radiation has been employed in organic and inorganic syntheses, oxidation/reduction reactions, and polymerizations [19].

Baek et al. [59] refer to the advantage of microwave radiation based on the high absorption of the GO compared to the solvent and metal oxide precursors. GO acts as the principal microwave absorber and can, therefore, be selectively heated, leading to the nucleation of the metal oxide onto its surface [59]. Generally, the precursor solution was ultrasonicated and magnetically stirred over a period of time. Afterwards, the slurry was placed in a microwave oven under cyclic microwave radiation for several cycles. Cyclic microwave radiation was employed in order to avoid bumping. The procedure continues with centrifugation and drying of the product [19,60]. Other previous works that use microwave approach include the preparation of Ag/GO nanocomposites [60], Mn_3O_4/rGO nanocomposites [19], GO/ $Ni_{0.4}Zn_{0.4}Co_{0.2}Fe_2O_4$ nanocomposites [62], and GO/ZnO [61].

2.8. Photocatalysis

Photocatalysis is also one of the techniques to fabricate GO-based material nanocomposites. Photocatalysis is applied as a synthesis method since the functional groups on the graphene sheet cannot be removed completely [63]. This one-step strategy can be utilized to fabricate high features of graphene based nanocomposites without using any toxic stabilizing reagent in the reduction reaction [64]. Besides, the prolonging of the photocatalytic reduction can be tuned on demand by controlling reaction time [63].

As is well known, GO consists of oxygen-containing functional groups including epoxy, hydroxyl, carbonyl, and carboxyl functional groups. Therefore, photocatalysts like ZnO and TiO_2 can strongly bond with these functional groups and readily distribute on the surface of GO, which also facilitates the photocatalytic reduction of the GO [63]. Under UV illumination, the photocatalyst will excite the electrons from valence band to conduction band and create electron-hole pairs, which can migrate and initiate redox reactions with water and oxygen [64]. Most of the oxygen-containing functional groups of GO are reduced and the photocatalyst particles remain on the surface of rGO via electrostatic and/or van der Waals forces [63]. Several studies have reported on the fabrication of GO-based material nanocomposite using photocatalytic synthesis such as GO/TiO_2 [63], GO/ZnO [64], GO/Ag [65], rGO/Pd [66] and rGO/porphyrin/Ag nanoparticles [67]. Based on the results of each study, photocatalytic synthesis is proven to fabricate the nanocomposite with an enhanced photocurrent generation and improved photocatalytic activity.

3. Application of Graphene Oxide-based Material Nanocomposite

3.1. Wastewater Treatment

Rapid industrialization has led to an increase in discharge wastewater containing heavy metals [68] and organic dyes [5]. Recent studies have reported photocatalysis [69], adsorption [70], advanced oxidation process [71], and sonocatalytic for the treatment of wastewater [72]. Various GO-based material nanocomposites have been studied that could make a large contribution to the wastewater treatment especially in heavy metal removal. In fact, GO and rGO are well known in photocatalyst and adsorption applications. The large specific surface area and abundant functional groups make GO a strong candidate for adsorption and photocatalyst applications in wastewater treatment.

3.1.1. Photocatalysis

There is great interest in the synthesis of graphene oxide nanocomposites for photocatalytic degradation of insecticides, organic dyes, heavy metals and other carcinogenic chemicals. Photocatalysts,such as TiO_2, ZnO, ZnS, and WO_3 are commonly employed because of the reusable and self-regenerated properties [40,69]. However, the practical purposes are narrow due to the rapid recombination of photogenerated electrons and holes within the photocatalyst [73]. Because of the excellent mechanical strength, low density, high catalytic activity, high surface area and the superior electron-transporting properties, graphene oxide can be exploited as a competent electron acceptor to boost the photoinduced charge transfer for amended photocatalytic activity [69,73].

Recently, there have been a few researchers who assessed the performance for photodegradation of organic dye like Rhodamine B and methylene blue by using graphene oxide-based material nanocomposites. Sun et al. [27] evaluated the addition of GO in the semiconductor with low loading of GO (0–0.5%) to enhance their photocatalytic activity under visible light irradiation. The photocatalytic activity of rGO/Cu_2O ($k = 7.85 \times 10^{-3}$) was reported exceeding that of pure Cu_2O and Degussa P25 by a factor of 2.9 and 7.9, respectively [27]. However, a further increase of graphene oxide content resulted in a gradual decrease in the photocatalytic activity due to the prevention of light from reaching the surface of the Cu_2O by the excessive GO, which would also shield the Cu_2O from absorbing visible light. Besides that, the loading of GO would inhibit the crystallization of Cu_2O that resulted in low photocatalytic activity.

Similar to previous work by Li and Hai [60], the rGO/Ag nanocomposite showed superior photocatalytic activity which was analysed by Rhodamine B as a model contaminant. However, as the microwave cyclic increases the photodegradation performance becomes weaker, which is attributed to serious oxidation of partially reduced GO. The result is supported by Choi et al. [74] who also observed a dwindling of the activity of rGO/CuI nanocomposites that perhaps due to the slight solubility of catalyst in aqueous solution. Hence, the microwave cyclic time is clearly substantial for the outcome on photodegradation of GO-based nanocomposites for Rhodamine B. The presence of graphene oxide also gives added value to a non-visible light photocatalyst, CuI into a visible light photocatalyst. CuI has a high band gap that has not allowed it to exhibit

photocatalytic activity in the visible region. After the hybridization excited electron from stimulated sunlight was injected from rGO nanosheets to the conduction band of CuI, it will generate hydroxyl and superoxide radicals that are sustained and enhanced by the more stable carbon free radical derived from rGO nanosheets, resulting a visible light activity [74].

In an attempt to optimize the parameter of the photocatalyst for Rhodamine B degradation, Maruthamani et al. [75] synthesized rGO/TiO$_2$ nanocomposites and studied the influence of rGO content, initial dye concentration, pH, and catalytic dose on decolorization. The result confirms that as rGO content in the composite was raised, more TiO$_2$ particles were able to attach onto the rGO sheet; therefore, extra charge carriers form exceedingly reactive species and boost the degradation of Rhodamine B. The percentage decolorization of RhB dye increased with the addition of the catalyst (up to 1.5 g L^{-1}) due to the surge in the number of active sites that promote hydroxyl radicals and enhance the dye adsorption on the catalyst surface. Then again, the percentage decoloration falls with a further increase in the catalyst (1.5 to 2 g L^{-1}), which may be ascribed to the agglomeration of the catalyst particles together with an increase in the turbidity of the suspension, which triggers light scattering. This results in decreased UV light penetration during the reaction [75].

Graphene oxide anchored to TiO$_2$ is one of the most common nanocomposites to be employed as a photocatalyst, as TiO$_2$ is an important candidate for photocatalytic decontamination. TiO$_2$ is well known to its high photocatalytic efficiency, chemical stability, antibacterial property, low toxicity, cost effectiveness, abundant and strong oxidizing material [76–78]. Yet, its high band gap energy (3.2 eV) contributes to low photoresponse toward sunlight, and the recombination effect becomes a drawback to its photocatalytic performance [76,77]. Thus, the remarkable electron capture-storage-transport properties of GO could enhance the charge separation efficiency of TiO$_2$ [79].

Gao et al. [78] modified the surface of water filtration polysulfone base membranes with GO/TiO$_2$ nanocomposite in order to enable photoactivity under both UV and sunlight. Four kinds of TiO$_2$ nanostructures were prepared including a 1D TiO$_2$ nanotube, 1D TiO$_2$ nanowire, 3D TiO$_2$ sphere assembled by nanoparticles (TiO$_2$ sphere-P) and 3D TiO$_2$ sphere constructed by nanosheets (TiO$_2$ sphere-S). The outcomes show that GO-TiO$_2$ sphere-S composites expressed photodegradation and disinfection activity superior to TiO$_2$ sphere-S under solar light irradiation. For this reason, optimizing the nanostructures of the photocatalyst coupled with carbon materials are two favorable attempts to improvise the photocatalyst [78].

Some researchers have also employed a ternary photocatalyst system such as rGO/TiO$_2$/ZnO [76], rGO/CoFe$_2$O$_4$/TiO$_2$[69], rGO/Ag/TiO$_2$/γ-Fe$_2$O$_3$[79], rGO/CdS/ZnO [80], and rGO/W/BiVO$_4$[81]. The competency of the ternary system was appraised and compared using a binary system (coupled with GO) and single photocatalyst material. The degradation competency of the ternary system was the best photocatalyst as confirmed by [76,79,80]. The surface area of the ternary system nanocomposite was also enhanced, which resulted in an increased adsorptivity, as reported by [69,79].

In a photocatalytic system, a reaction takes place at the surface of the catalyst. The generation of an electron-hole pair plays a significant role in the mechanism of a photocatalytic reaction. When a photocatalyst is exposed by a light stronger than its band gap energy, electron-hole pairs diffuse out to the surface of the photocatalyst and participate in a chemical reaction with the electron donor and acceptor [40]. Here, the valance band (VB) electrons (e$^-$) of the photocatalyst are excited to the conduction band (CB), creating holes (h$^+$) in the VB [27]. Those free electrons and holes transform the surrounding oxygen or water molecules into hydroxyuracil (OHU) free radicals with super strong oxidation [40]. These free radicals are then used to decompose the organic pollutant into carbon dioxide and water [77]. A highly efficient visible light photocatalysis should have high quantum efficiency resulting from low recombination of the photogenerated electron-hole pair and a wide light response range because of the narrow band gap [27]. With the presence of excellent electron-mobility of GO anchored to a photocatalyst, the charge transport rate could be increased, therefore, inhibiting the charge recombination and promoting the photocatalytic activity, consequently, through a quick electron-hole transfer [60]. The presence of GO also confirmed that it could reduce the band gap that promotes an excellent photocatalytic activity [75]. The list of GO/rGO-based material photocatalysts with the material detection is shown in Table 1.

3.1.2. Adsorption

Many GO-based composites for adsorption of contaminants from wastewater have recently been developed. The magnetic composite, like Fe$_3$O$_4$, is one of the promising materials for the wastewater

remediation. Boruah et al. [70] systematically synthesized Fe_3O_4 with rGO for methylene blue adsorption with an adsorption capacity of 75.15 mmol/g, which is higher than Fe_3O_4. The results are supported by Hao, Wang, Gou, and Dong, [84] who fabricated GO/Fe_3O_4 as an adsorbent for removal of Chrysoidine Y. GO is exfoliated by strong oxidants, and it consists of a hexagonal network of covalently linked carbon atoms attached with oxygen-containing functional groups, such as epoxy, carboxyl and hydroxyl groups at various sites [1].

Table 1: The list of photocatalysts and material detection in previous study.

Photocatalyst	Material detection	Researcher
rGO/Cu_2O	Rhodamine B	[27]
rGO/Ag	Rhodamine B	[60]
rGO/CuI	Rhodamine B	[74]
rGO/TiO_2	Rhodamine B	[75]
rGO/TiO_2	Bisphenol A	[23]
$rGO/Ag/TiO_2$	Methylene blue	[82]
$rGO/Ag/TiO_2/\gamma\text{-}Fe_2O_3$	Crystal violet	[79]
$rGO/CoFe_2O_4/TiO_2$	chlorpyrifos	[69]
GO/TiO_2	*Escherichia coli (E. coli)*	[78]
GO/TiO_2	Methylene blue	[77]
$rGO/TiO_2/ZnO$	Methylene blue	[76]
rGO/ZnO	Methylene blue	[40]
rGO/CdS/ZnO	Methylene blue	[80]
$rGO/W/BiVO_4$	Methylene blue	[81]
rGO/Pd/Ni	4-chlorophenol	[83]

Anchoring Fe_3O_4 nanoparticles onto GO might conjoin the relatively high adsorption capacity of graphene oxide and the separation convenience of magnetic materials, which can bind with cationic dyes through the π-π stacking interaction and van der Waals forces [84]. In a similar work, Wang et al. [68] evaluated the adsorption performance of ternary magnetic composites consisting of rGO, polypyrrole and Fe_3O_4 nanoparticles for the Cr(VI) removal with adsorption of 293.3 mg/g, which is much higher compared to rGO/Fe_3O_4 nanocomposites. Magnetic graphene based adsorbent that assists separation by the magnetic field can be used since it is hard to separate the small particle size of GO from an aqueous solution through a conventional centrifugation and filtration method [68].

Polypyrrole along with GO is widely used for Cr(VI) removal, as polypyrrole has high chemical stability, ion exchange ability, ease of preparation, and low cost [6,85]. The existence of polypyrrole enhances the adsorption ability due to positively charge nitrogen atoms that easily bind heavy metal [68]. The hybridization between polypyrrole, GO and α-cyclodextrin make a perfect adsorbent with a strong acid resistance and superior mechanical properties because it has a remarkably larger surface area, abundant hydroxyl groups and cavities [6].

The Cr(VI) removal is mostly via electrostatic attraction; also, some might involve ion exchange and chemical reduction processes [68]. A previous study done by Setshedi et al. [85] affirmed that a drop in Cr(VI) removal efficiency is influenced by increasing the solution pH. At lower pH values, greater Cr(VI) removal efficiency could be achieved as the surface of nanocomposites is positively charged due to the detachment of doped Cl^- ions with simultaneous protonation of the nitrogen atom in the presence of sufficient H^+ ions. This result was strongly supported by Wang et al. [68] and Dong et al. [86] who stress that the decreasing adsorption capacity was due to higher pH. Their result is also supported by zeta potential values, which decrease with an increase in the solution pH, due to nitrogen groups' deprotonated [68,86,87]. However, Liu et al. [5] conveyed an opposite opinion, which proved that as the solution pH increases, the adsorption capacity will also increase. This is because the contaminant is in a cationic form, and lower pH will provide insufficient electrostatic attraction between the cationic contaminant and the nanocomposite [5,88]. So at lower pH, positively charged becomes totally dominant at the adsorbent surface, resulting in an excellent electrostatic attraction between the anionic contaminant and vice versa.

Sheshmani et al. [87] also used the cationic contaminant (Pb(II) ion), yet the adsorption became weaker as the pH increased. The interactions take place between the metal ion and the amino groups of the nanocomposite. At low pH, H^+ and Pb^{2+} are competitively adsorbed on the surface of the adsorbent because the

amino group of GO/chitosan/FeOOH nanocomposite can be simply protonated to be -NH$_3$. This reduces the adsorption capacity as there is an increase of electrostatic repulsion between -NH$_3$ and Pb^{2+}. At higher pH, the protonated -NH$_3$ decreases, which increases the recovery of -NH$_2$ resulting in an increase of the amount of adsorbed Pb^{2+}. However, with a further increase in pH, OH$^-$ can be adsorbed onto the surface of -NH$_2$ competing with Pb^{2+}, leading to a reduction in the amount of adsorbed Pb^{2+}[87].

The surface area is also a significant factor in adsorption. Setshedi et al. [85] affirm that the increased Cr(VI) sorption efficiency of the GO/polypyrrole may be due to the increased surface area. Conversely, Wang et al. [68] show that the surface area is not a key factor for rGO/polypyrrole/Fe$_3$O$_4$ nanocomposite to be a good adsorbent. Therefore, there are three possible opinions; first, after the hybridization, graphene oxide might help increase the surface area of the adsorbent material and lead to an increase in adsorption capacity [85,86]; second, the adsorbent material itself helps increase the surface area of graphene oxide as GO is poorly dispersed in water and lead to a decrease its surface area [5]; and lastly, the hybridization of GO with adsorbent material did not influence the surface area at all but the adsorbent still has an excellent adsorption performance [68].

The affinity of GO is already acknowledged as the main factor of a good adsorbent. The affinity can be assigned to the oxygen-containing functional groups and the aromatic matrix [89]. The oxygen-containing functional group tends to bind to the hydrophilic species owing to their electrostatic interactions or hydrogen bonds, whereas the aromatic matrix is likely to bind to the hydrophobic organics by π-π stacking or hydrophobic interactions [90]. However, the relative ratio of the two functional regions in GO is adaptable with oxidation conditions, and offers GO with a range of adsorption selectivity and characteristics [91]. Thangavel and Venugopal [92] proved that as the oxidation level of GO increases, the adsorption capacity increases. This is due to the extra number of functional groups that were introduced onto the basal plane of GO through the oxidation process, and the increase in the negative charge of the GO sheets [89,92]. Furthermore, some of the anionic groups on GO will also be deprotonized at higher pH resulting in an enhanced negative charge [89].

Yan et al. [89] has found that adsorption behavior of GO towards methylene blue changes from a Freundlich-isotherm to a Langmuir-isotherm adsorption as the oxidation degree increases. At a lower oxidation degree of GO the isotherm behavior follows the Freundlich model, which reveals a multilayer adsorbtion on sorbent [93]. At a higher oxidation degree of GO, however, the isotherm behavior follows the Langmuir model, which is applied on the single layer adsorption mechanics on the surface of the adsorbent [93]. The binding of a lower oxidation degree of GO toward methylene blue is mainly through parallel π-π stacking interactions and form multilayer adsorption, while GO with a higher oxidation degree shows monolayer adsorption through electrostatic interactions [89].

A GO-based nanocomposite could be a first-class preference as an adsorbent for adsorbing assorted organic molecules, because of its large specific surface and reactivity. Moreover, GO is capable of forming strong π-stacking interaction with a benzene ring because its greater delocalised π-electron system fabricates it for adsorption of aromatic compounds.For comparison, the material detection, maximum adsorption capacity, and pH of these GO-based nanocomposites are summarized in Table 2.

3.1.3. Advanced Oxidation Process/Sonocatalytic

In a broad sense, the advanced oxidation process (AOP) is an established chemical treatment procedure invented to remove organic and inorganic materials in water and wastewater by oxidation through reactions with hydroxyl radicals.Current trends are the development of modern and modified AOPs that are efficient and economical, such as the implementation of ultrasonic treatment or also known as sonocatalysis [95].

Presently, sonocatalytic waste water treatment has obtained remarkable attention from researchers. This is an alternative method to the photocatalytic degradation [96]. With a high demand on an excellent catalyst that can be regenerated in dye removal efficiency, this method can fulfill the criteria that can decrease the negative effects of dye effluents [97]. Their remarkable efficiency without additional oxidants can reduce the treatment expense [98]. Sonocatalytic technology implies the use of ultrasound as a source of high energy at a frequency range of 18–100 kHz to initiate the formation of acoustic cavitations. It comprises the formation, growth and collapse of cavity bubbles that entangles dissolved vapors surrounding water[47,97]. Several studies on sonocatalytic degradations have shown higher efficiency compared to photocatalytic degradation [72]. Zhu et al. [98] reveal that ZnSe-GO/TiO$_2$ sonocatalyst is used to accelerate the degradation of organic azo through acoustic cavitation. It proves that sonocatalysis is significant for initiation catalytic reaction in both homogeneous and heterogeneous cases [98].

Table 2: List of adsorbent, material detection, and maximum adsorption capacity from previous study.

Adsorbent	Material detection	Maximum adsorption capacity, mg/g	pH	Researcher
GO/Fe_3O_4	Chrysoidine Y	344.83	7	[84]
$GO/porphyrin/Fe_3O_4$	sulfonamides	13.9-sulfanilamide 10.9sulfadimidine 44.2-sulfadimethoxine 28.3-sulfadiazine 10.5-sulfaguanidine 36.3-sulfamethoxazole 26.6-sulfapyridine	4	[94]
rGO/Fe_3O_4/polypyrrole	Cr(VI)	293.3	3	[68]
GO/polypyrrole	Cr(VI)	625	2	[85]
GO/α-cyclodextrin/polypyrrole	Cr(VI)	606.06-666.67	2	[6]
GO/β-cyclodextrin/poly(acrylic acid)	Methylene blue (MB) & safranine T (ST)	247.99-MB 175.49-ST	98	[5]
GO/schwertmannite	Sb(V)	158.6	7	[86]
GO/chitosan/FeOOH	Pb(II)	111.11	5	[87]

In this study, the presence of TiO_2 as a photocatalyst has enhanced the sonocatalytic decomposition of Rhodamine. Similar work has been done by Zhu et al. [96]which shows that the capability of the GO-TiO_2 composites to adsorb Rhodamine, owing to the formation of π-π stacking between Rhodamine molecules and noncovalent aromatic regions of graphene. The oxidation process of dyes is reliant on OH·. The heterogeneous nucleation of bubbles can increase the formation of cavitation bubbles, then generating hot spots in the solution. These hot spots pyrolyze H_2O to form OH·. Subsequently sonoluminescence, which involves intense UV light, excites the TiO_2 particles to act as photocatalysts during sonication. The combination of photocatalyst particles and ultrasound treatment can offer an extra number of nuclei for cavitation bubble formation [98]. Degradation of organic compounds involves the sonolysis of water as the solvent inside the collapsing cavitation bubbles under extremely high temperatures and pressure. With the addition of a catalyst, ultrasonic irradiation not only induces sonolysis of water, but also couples with the catalyst to produce electron-hole pairs. The electron-hole pairs can produce OH· radicals and superoxide anions $\cdot O_2^-$, which can decompose dyes to CO_2, H_2O, and inorganic species [96,98].

Through the study carried out by Thangavel et al. [72], it was shown that the degradation rate of GO/Fe^{3+} sonocatalyst is enhanced when electron scavengers like peroxomonosulphate, peroxodisulphate, hydrogen peroxide and potassium periodate were added, while the degradation decreases with the addition of inorganic ions like SO_4^{2-}, Cl^-, $H_2PO_4^-$, and HCO_3^-. The promotion of degradation by electron scavengers occurs when the electron scavengers react with Fe^{2+} then generate high radical species. Additionally, the adsorption process is reduced as the inorganic ion covers the surface of the $GO-Fe^{3+}$ hybrids system due to the electrostatic interaction, and inorganic ions react with Fe^{2+}-Fe^{4+} to produce the species less reactive than OH·[72].

3.1. Electronic

There are many electronic devices that have been invented using GO as a starting material, such as a supercapacitor and lithium ion batteries. A supercapacitor and lithium ion battery are presumed to be an excellent choice for energy storage [17,19,20,26,99]. Various metal oxides have been employed to develop these devices. However, these metal oxides have certain drawbacks that interfere with the chemical and technical process. Therefore, GO and rGO are hybridized with these metal oxides in order to increase the performance of the device.

3.2.1. Supercapacitor

A supercapacitor, also known as an electrochemical capacitor, is an energy storing device that has attracted much attention in recent years. Due to its advantages, such as high power density, long life cycle and fast charging/discharging rates, a supercapacitor is also used as an intermediate system between dielectric capacitors and batteries [18] as well as conversion of electric devices [100]. The main challenges of the existence supercapacitor are the lower energy density and poor overall performance [14]. Therefore, the GO-based nanocomposite is introduced as a new supercapacitor with enhanced physicochemical properties, such as outstanding electronic conductivity, superior mechanical strength, and greater surface area [99].

The presence of GO and rGO can increase the surface area of the nanocomposite. Work done by Ji et al. [99] affirms that rGO can inhibit agglomeration and manipulate the ceria's structure. It is commonly acknowledged that the smaller particles have a better propensity to aggregate and reduce the surface area, specifically in nanoscale. That theory was supported by surface morphology studies that show that the cascading structure of supercapacitor material and GO morphology assists in an increase in the specific surface area of the composites and boosts electrical conductivity [17]. The list of various GO/rGO based-material nanocomposites fabricated as supercapacitor material is shown in Table 3.

Table 3: The specific capacitance and capacitance retention of different types of nanocomposite in supercapacitor application.

Nanocomposite	Specific capacitance, F/g	Capacitance retention	Researcher
GO/polyaniline	531.0	-	[18]
GO/manganese dioxide	216.0	84.1 % after 1000 cycles	[101]
GO/polyaniline	355.2	80.46 % after 1000 cycles	[17]
GO/cobalt tungstate	159.9	94.7 % after 1000 cycles	[14]
rGO/polypyrrole/sodium dodecyl benzenesulfonate	277.0	50 % after 500 cycles	[35]
rGO/boron nitride	140.0	105.5 % after 1000 cycles	[100]
rGO/polyaniline	701.0	92 % after 1000 cycles	[37]
rGO/cerium(IV)oxide	265.0	96.2 % after 1000 cycles	[99]
rGO/manganese(II,III)oxide	160.0	91.5 % after 800 cycles	[19]

3.2.2. Lithium ion battery

The lithium ion battery has become one of the most widely used secondary batteries for portable electronics, hybrid electric vehicles and renewable energy storage [102]. Electrochemically active metals and metal oxides such as CuO [103], $TiOF_2$ [104], Fe_3O_4[105],CoF_2[106], $CdWO_4$[107], SnO_2 [108], TiO_2[109], and Mn_2O_3[110] have long been considered as anode materials for lithium ion batteries because of their remarkable high theoretical capacities [111]. All of these materials become attractive candidates for electrode material of lithium ion batteries because they are environmental friendly, and have structural stability and minimum cost for production [112–114].

Despise these transcendent properties, there are a few drawbacks that a researcher must face. Taking TiO_2 as an example, it bears a low Li-ion diffusivity, theoretic capacity, and low electronic conductivity in reversible Li insertion/extraction processes [63,113]. Similar to TiO_2, one of the most recognized semiconductors, it also has its downsides. The electrical conductivity CuO in the charge state is relatively poor and it has a large volumetric change of the electrode that can damage its crystal structure and affect its capacity [114]. So GO and rGO is assigned to support the mechanical and electrical properties of the existing material.

Peng et al. [102] has hybridized rGO with selenium nanoparticles in order to maximize the capacity and energy density of the lithium-selenium battery cathode. Furthermore, the rGO framework also helps to reduce the selenium loss and prevent the polyselenides from leaving during cycling [102]. The presence of these carbon based materials has also been confirmed to contribute to the long-term cycle stability, excellent energy capacity, enhanced electrochemical performance, and large reversible capacity compared to the pristine materials [20,26,112,114]. The list of GO/rGO based-material nanocomposites that already have been fabricated as a supercapacitor material is shown in Table 4.

Table 4: The nanocomposite assigned as supercapacitor in previous study.

Materials	Initial discharge capacity (mA h g^{-1})	Reversible capacity (mA h g^{-1})-cycles	Current density (mA g^{-1})	Researcher
rGO/selenium	533	265-500	675	[102]
rGO/TiO$_2$	368	136.1-100	1000	[113]
rGO/titanium dioxide	295.4	112.3-100	1000	[26]
rGO/MnO	1017	988.6-120	100	[20]
rGO/TiO$_2$	310	270-100	100	[63]
GO/CuO	1369.3	590-50	100	[114]

3.3. Self-cleaning

Recently, self-cleaning technology has begun to grow rapidly since self-cleaning coatings and windows have a huge commercialization demand. Self-cleaning materials should have good photocatalytic activity with high durability, optical transparency for coating surface, and superhydrophilicity properties [22]. Superhydrophilicity is a state of material having strong affinity to water greater to non-polar air, which is a very important property needed for a self-cleaning coating [115].

Thakur and Karak [116] have fabricated a rGO/TiO$_2$ nanocomposite that shows excellent self-cleaning properties. The report revealed that the self-cleaning properties increase with an increasing amount of TiO$_2$. The increasing amount of rGO, however, will assist in effective healing properties [116]. He [22] shared the same viewpoint, which is that the photocatalytic activity improved with an increase of the rGO/ZnO ratio, due to three factors; first, the development of their charge separation due to electrons infusion from the conduction band of ZnO to graphene; secondly, the reduction of average particle size; and third, the increase in superhydrophilicity.

In addition, Yun et al. [117] reported that a prepared GO/TiO$_2$ nanocomposite had excellent self-cleaning properties toward methylene blue stained films. Similar to the work done by Thakur and Karak [116], the increasing content of GO led to an improvement in film transmittance and higher efficiency in methylene blue adsorptivity. With superhydrophilicity of GO, it can be a suitable candidate for both an adsorbent and a binder [117]. However, GO also can be assigned to be superhydrophobic coating to lower the moisture adsorption and enhance the reliability of the electronic device. Lin et al. [7] have fabricated GO as a superhydrophobic coating material, which is functionalized by aliphatic amine. These aliphatic amine molecules consist of a hydrophobic tail and hydrophilic head anchored onto the GO by a nucleophilic reaction between the amine group and epoxide group [7]. This study illustrates that GO surface functionalization can lead to various functional applications of GO films.

3.4. Sensor

Sensors based on graphene oxide-based nanocomposite material are getting endless attention due to their ultrafast response [9], great sensitivity [25], their esteemed long-term stability [118], superb conductivity [36], reproducibility [119] and facile fabrication [50,119–121]. Recently, nanocarbon material has appeared to be a momentous perspective in the topic of humidity and gas sensor. Its sensitivity towards water molecules is due to the numerous oxygen functional groups that are decorated on the basal plane and the edge of graphene oxide, such as carboxylic acid, hydroxyl, and epoxy groups, which can expand the hydrophilicity [9]. Nanostructure material normally shows certain apparent benefits for sensor purposes, for instance, tremendous adsorption capacity, large specific surface area, great mechanical stiffness, extraordinary high carrier mobility and better stability [9,122]. So far, many researchers have employed GO-based material to detect humidity [9,122], hydrogen peroxide [25], trimethylamine [29], ammonia [123], and nitrogen dioxide [118].

To date, many biosensors based on GO-based nanomaterial have caught researchers' attention. One of them is the glucose biosensor, which is successfully synthesized by one step electrodeposition [32]. In this study, rGO/β-lactoglobulin acts as a stabilizer and an excellent template for the growth of dendritic gold nanostructures (Au Nps). Au Nps draws a particular consideration in the electrochemical field since it has good chemical stability and a large surface area, is exceptionally biocompatible, enhances the electrode conductivity and is capable of assisting electron transfer between electrodes and biomolecules [50,124]. In a study done by Peng et al. [50], Au NPs were employed to immobilize the probe DNA via Au-S bond to detect a multidrug

resistance gene in cancer treatment. The hybridization of GO with toluidine blue and Au NPs, GO was used to enlarge the loading capacity of the toluidine blue and preclude the leakage of the toluidine blue from the electrode surface [50]. Yola et al. [124] also employed GO anchored to Au involving 2-aminoethanethiol for detection of tyrosine in milk, which makes a superior sensor with a low detection limit [124]. The list of GO/rGO based-material nanocomposite sensors, material detection, and detection limit is shown in Table 5.

Table 5: The type, material detection and detection limit of previous synthesized sensors.

Type of sensor	Sensor	Material detection	Detection limit	Researcher
Gas sensor	GO/poly(diallyldimethylammonium chloride)	Humidity	-	[9]
	rGO/Ag	Hydrogen peroxide	0.9×10^{-6}M	[25]
	GO/nanodiamond	Humidity	-	[122]
	GO/WO$_3$	Triethylamine	-	[29]
	rGO/polypyrrole	Ammonia	-	[123]
	rGO/WO$_3$	NO$_2$	-	[118]
Biosensor	rGO/β-lactoglobulin/Au	Glucose	2.29×10^{-5} M	[32]
	GO/Au/toluidine blue	Multidrug resistance	2.95×10^{-12}M	[50]
	rGO/ZnS/	Ascorbic acid (AA), dopamine (DA) and uric acid (UA)	3×10^{-5} M-AA 5×10^{-7} M-DA 4×10^{-7} M-UA	[120]
	GO/Pt/CeO$_2$/1-naphthol	Influenza	4.3×10^{-13}g mL^{-1}	[125]
	GO/multi-walled carbon nanotube	Sunset Yellow (SY) and Tartrazine (TT)	2.5×10^{-8}M-SY 1×10^{-8} M-TT	[126]
	rGO/Fe$_3$O$_4$	Acetylcholine	3.9×10^{-8} M	[121]
	rGO/CeO$_2$	NO	9.6×10^{-9} M	[127]
	rGO/chitosan/tyrosinase	Bisphenol A	7.4×10^{-10} M	[33]
	rGO/SnO$_2$	Dopamine	1×10^{-6} M	[128]
	GO/Au/2-aminoethanethiol	Tyrosine in milk	1.5×10^{-10} M	[124]
Environmental sensor	rGO/poly(3-aminophenylboronic acid)	Fluoride	9×10^{-11} M	[34]
	rGO/polyaniline	Mercury ion	3.5×10^{-11} M	[36]
	GO/Ag	Nitrite ion	2.1×10^{-6} M and 3.7×10^{-5} M	[119]
	GO/Ag	Dye	1×10^{-6} M	[129]

3.5. Catalyst

Various catalysts based on the GO nanocomposite have been studied recently. Despite the fact that there has been a significant effort committed to the utilization of different metals as a suitable catalyst, there is still a need to find suitable backings for the catalyst framework; this zone needs to be investigated more thoroughly.

One of the latest studies was done by Zahed and Monfared [130] who synthesized GO/Ag nanocomposite as a catalyst for aerobic oxidation of benzyl alcohol. Silver is well known as a catalyst for many oxidation reactions. The Ag particles that consist of very small particles can contribute to a greater specific surface area, but it also might lead to agglomeration from corrosive surface energy. Therefore, the presence of GO can support and separate the nanoparticle, preventing it from agglomerating and making it recyclable [130]. This catalyst is also confirmed to be reused several times. In different works, GO/Ag nanocomposite is

employed as a catalyst for reduction of 4-nitrophenol [131]. This nanocomposite was reported to be highly reactive and a very stable catalyst. The high activity of the GO/Ag nanocomposite may be credited to presence of GO, which also leads to exceptionally productive contact between 4-nitrophenol and Ag nanoparticles on the GO surface.

In a previous work done by Song et al. [24], GO-COOH has inherent peroxidase-like action and its catalysis is emphatically subject to pH, temperature, and H_2O_2 concentration, like horseradish peroxidase. Peroxidase has extraordinary potential for handy applications and can be utilized as a diagnostic kit for hydrogen peroxide and glucose [24]. Horseradish peroxidase has been generally used to manufacture sensors for product detection of the glucose oxidase. GO-COOH is low cost, simple to acquire, steadier to biodegrade, and less defenseless against denaturation compared to horseradish peroxidase. Hence, these aspects show that GO-COOH can be valuable in ecological observations and medicinal diagnoses. A few of the recently fabricated GO/rGO based-material nanocomposite catalysts are listed in Table 6.

3.6.   Biomedical

A graphene oxide-based nanocomposite has exposed fascinating purposes in the biomedical field. GO is extensively used in biomedical applications because its two-dimensional plane and one-atom thickness give it superior specific surface area for immobilization of numerous substances, including a wide range of metals, biomolecules, fluorescent molecules and drugs [134].

Yang et al. [134] has reported a hybrid of GO with superparamagnetic Fe_3O_4 for controlled targeted drug carrier The GO/Fe_3O_4/doxorubicin nanocomposite exhibits a hydrophilic surface and its superparamagnetic properties make it congregate easily in an acidic environment and move in a magnetic field; moreover, it can simply be redispersed to make a stable suspension under basic conditions [134]. Similarly, Zhang et al.[135] have also fabricated GO as a novel nanocarrier for the loading and targeted deliveries of anticancer drugs by functionalize it with sulfonic acid and binding it with folic acid. Compared to nanocomposite synthesized by Yang et al. [134], this nanocomposite tends to load two anticancer drugs, which are doxorubicin and camptothecin simultaneously. Since they have an efficient loading property of multiple anticancer drugs, the therapeutic efficacy could be enhanced, which leads to wide potential clinical practice [135].

Table 6: The previously synthesised catalysts and their catalytic activity.

Catalyst	Catalytic activity	Researcher
rGO/Cu	Reduction of 4-nitrophenol	[132]
GO/Ag	Reduction of 4-nitrophenol	[131]
GO/magnetite/Ag	Reduction of 4-nitrophenol	[133]
GO-COOH	Reduction of H_2O_2	[24]
GO/Ag	Aerobic oxidation of benzyl alcohol	[130]

Titanium and its alloy is well known as electrochemical devices due to the large surface area, excellent mechanical properties, bioactivity and corrosion resistance [136]. With that, Yan et al. [136] have employed GO cross-linked gelatin as reinforcement fillers in a hydroxyapatite coating by an electrochemical deposition process on TiO_2 nanotube arrays. The enlarged surface area of the nanotubular surface of TiO_2 offers huge active reaction sites for chemical reaction, and develops the growth, adhesion, and differentiation of the cell [136]. Additionally, Wang et al. [137] have explored the sensing property of GO/aptamer-carboxyfluorescein in vitro and in situ molecular probing in living cells. The effectual release to molecular targets in living cells shows that GO could be a decent vehicle to transport genes into cells, shielding the loading genes from the enzymatic cleavage and empowering in situ molecular probing in living cells [137].

4.   The Newly Hybridized Graphene-like Derivatives in Advanced Photocatalytic, Energy and Environmental Remedies

Since the term graphene was first used, much research has been proposed and conducted on graphene-like derivatives. Quite a few allotropes of carbon such as graphyne, graphane, graphdiyne, and fluorographene, have come about, which are deemed as two dimensional analogue materials to graphene [138,139]. Among these

materials, graphdiyne (GD) seems to catch a lot of interest owing to its optical, structural, and emergent electronic properties, which are distinctive from other carbon variants. GD cannot be synthesized directly from graphene, but they are compared and discussed with graphene with respect to their structure and properties [139]. Its structure involves two acetylenics in-between carbon hexagons in repeating order, and constructed by $sp-$ and sp^2-hybridized periodic carbon networks [140].

Owing to its unique properties, GD is a promising material for electronic and energy applications. GD has the potential to be employed as a semiconductor due to its low effective electron mass and the band gap. When the effective electron mass is so low, the electrons should move very quickly, and even a slight potential field is used [141]. In a previous work done by Gong et al. [142], the electronic structure and field emission properties of the GD-boron nitride composite has been investigated. This composite possesses an excellent stability due to the increase of its binding energy. The result of the shifting of HOMO and LUMO to vacuum level, the decrease of band gap with the increasing electric field, and an effective change in the local electron density distribution make this composite a promising candidate for field emission devices in the future.

In a recent study, GD hybridized ZnO showed a superior photocatalytic activity compared to GO/ZnO and rGO/ZnO photocatalyst [138]. The inclusion of GD in the prepared GD/ZnO photocatalyst decreases the absorption edge of ZnO, thus developing additional photon energy, which results in an enhanced photocatalytic reaction rate. Zhang et al. [140] has shown that the pristine GD species could be easily hybridized with Ag/AgBr. With GO acting as a dual-functional coupling reagent, this hybrid material could work as highly efficient visible-light-driven photocatalysts toward the photodegradation of methyl orange pollutant [140]. Being an excellent photocatalyst, the hybridization of photocatalyst particles with GD has potential in wastewater treatment, and can contribute to a remedy for environmental pollution. In a future work, GD is strongly proposed to be modified with metal oxide, semiconductor, photocatalyst particles and complex oxide in order to reach its full potential.

5. The Challenges Facing Graphene Commercialisation

While graphene may have garnered huge public interest, it remains to be seen exactly how much impact it will have. The commercialisation of graphene composites faces a major predicament. The biggest challenge to the commercial usage of graphene is the production cost, especially when competing with existing materials. Secondly, the manufacture cannot find the most required applications of graphene and its proper use that demands a large quantity of graphene. Maintaining close collaboration with downstream application companies is also one of the challenges for the development of future applications. Limited material availability can potentially slow down the commercial adoption of graphene.

In addition, the challenge lies in maintaining the quality in large quantities of graphene. If even a small defect is detected on the graphene monolayer carbon network, it will influence the electrical conductivity, the transparency, the impermeability, the thermal conductivity and all the other unique properties that are specific to graphene. There are only a small number graphene-based products that have reached the market, such as a tennis racket by Head, a battery strap by Vorbeck, an oil-drilling mud by Nanochem and a phone touch screen by Samsung [143]. Graphene manufacturers also need to be very careful when expanding their production scale, to ensure that there are appropriate uses for the material.

One of graphene's special properties is that it is highly conductive. Theoretically, this property is good for an electronic device. However, in an actual situation, it is too conductive for many applications in electronics, as it has no band gap. Countless studies have been done to introduce artificial band gaps into graphene, from patterning it into nanoscale ribbons to doping its surface with chemicals. Then again, this issue is stuck with the expensive production cost, as these methods are typically complex and expensive, making it difficult to adapt them for large-scale use in industry.

Its seems that the research efforts come to a dead end when production costs are involved. However, with the manufacturing side of the graphene industry continuing, research on integrating graphene into applications will be successful and graphene's future still looks very bright.

6. Conclusion

In summary, recent studies on graphene oxide-based nanocomposites are increasing rapidly. Because of the nanocomposites' spectacular and unique properties, researchers have been competing to develop a novel nanocomposite with various synthesis methods. The synthesis method plays a vital role in order to produce a larger scale production and to control the quality of the nanocomposite. At the same time, the methods should be

efficient, affordable, and safe for the environment. The presence of GO also makes a great contribution toward enhancing the performance of other material. Therefore, there is much work that needs to be done in order to manipulate the two-dimensional GO sheets for future advanced technology.

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