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Magnetic graphene oxide: Synthesis techniques and applications in gas-sensor and biosensors

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Abstract

In recent years, there has been a notable increase in interest in magnetic graphene oxide (MGO) due to its unique properties and potential applications in gas and biosensors. An extensive summary of the MGO synthesis techniques, such as chemical reduction, hydrothermal synthesis, and solvothermal synthesis, is given in this review article. Along with its many uses in gas and biosensors, sensitivity, selectivity, and stability of MGO are also highlighted. In addition to being a gas sensor that can identify ammonia, hydrogen sulfide, and volatile organic compounds, MGO can be used as a biosensor to identify proteins, glucose, cholesterol, and DNA. The conclusion of article discusses the future directions of the field as well as possible applications for MGO research across a range of industries.

Keywords: *Magnetic graphene oxide, Fe_3O_4/GO, Biosensor, Gas-Sensor, Sensor Mechanism*

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1. Introduction

Graphite can be treated with strong oxidizers to produce graphite oxide, which is a mixture of carbon, oxygen, and hydrogen in different proportions. Graphite oxide was originally known as graphitic oxide, or graphitic acid. At a C:O ratio of 2.1 to 2.9, the larger, unevenly spaced layer structure of graphite is retained. The substance forms monomolecular sheets, or graphite in a single layer, when it dissolves in basic solutions. This substance is also referred to as graphene oxide. Graphene oxide is hydrophilic by nature and comprises a range of reactive groups containing oxygen, such as carboxylic, hydroxyl, epoxy, and carbonyl groups. It shares the same general structure as graphene, but it is more bioactive. [1, 2]. Graphene oxide (GO) nanosheets are produced by chemically oxidizing bulk graphite particles [3, 4]. Extensive research has been conducted on GO and its derivatives in relation to a wide range of biodevice applications, including drug delivery, nanomedicine, bioengineering, biotechnology, biosensors, cell imaging, energy storage, and gas sensing [5, 6].

Extracting graphene oxide (GO) from aqueous solutions is challenging due to the limited direct applicability of graphene oxide nanosheets in solid-phase extraction and their propensity to clump in the aqueous phase [6, 7]. The best remedy for this problem is magnetic graphene oxide, a substance that blends graphene oxide with iron oxides. MGO nanocomposites may be able to be separated by applying an external magnetic field. However, in biomedical applications, the inherent cytotoxicity of GO and its derivatives needs to be considered as well [8, 9]. More research using different types of iron oxide nanoparticles has been conducted recently. Among these iron oxides, g-Fe_2O_3 and Fe_3O_4 nanoparticles are currently the most promising candidates for use in biomedical and sensor applications due to their remarkable biocompatibility, robust chemical stability, and lack of obvious intrinsic toxicity [10-12]. Interactions between an external magnetic field and iron oxide nanoparticles change the energy of system. These interactions may provide a solid foundation for the diverse applications of iron oxide nanoparticles. Since MGO is a composite of graphene oxide and iron oxide, it has the better physical and chemical characteristics of both [13, 14]. Among these qualities are para-magnetism, a high activity of surface-binding sites, a good surface area that is specific to oxidation, size coordination, strong chemical stability, simplicity in functionalization, and biocompatibility. By combining magnetic metal oxide, several researchers have produced graphene oxide-based MGO. Fe_3O_4/GO [15], magnetic reduced GO [16, 17], and other hybrid nanocomposites [14] are a few examples of these. Many researchers have looked into MGO in the areas of drug loading [18, 19], chemical extraction and separation [20, 21], magnetothermal therapy of tumors [22], photodynamic therapy [23, 24] and bio-imaging [25]. The primary goal of this paper was to present an overview of MGO use strategies for relevant biological and environmental applications. First, we go over how to prepare MGO nanocomposites for use in sensors. We will go into great depth about the physicochemical and structural functions of MGO nanocomposites as well as the potential cytotoxicity of using them in biomedical settings. Lastly, we address the challenges we currently face and make predictions about possible future research directions for the study of MGO nanocomposites for environmental applications.

The aim of this review paper is to demonstrate the possible applications of GO in the gas and biosensor industries. While MGO biosensors have been evaluated before, this review adds some new data and updates the most recent review. We enumerate the current uses of MGO as sensors and elucidate the basic

principles underlying detection methods. In conclusion, we offer an examination of the subject matter grounded in insights gained during the previous ten years, specifically the previous five.

2. Synthesis and structure of graphene

Chemical and physical processes are the two primary ways for creating MGO particles. Chemical synthesis techniques are used more frequently than the others. Chemical co-precipitation, solvothermal, hydrothermal, covalent bonding, in situ chemical methods, and electrochemical methods are the main categories of chemical methods. The ball mill method is one of the primary physical methods. The most widely used of these are the hydrothermal, solvothermal, and chemical co-precipitation processes; their synthesis can be regulated in mild circumstances [26].

2.1 Chemical co-precipitation

In summary, adding a specific ratio of Fe^{3+}/Fe^{2+} to the GO solution is a process of making MGO composites. An inert gas is then used to complete the remaining carboxyl and cationic interactions on the GO surface. The size and shape of magnetic nanoparticles of co-precipitation method can be influenced by several variables, including pH and reaction time [27, 28]. Magnetite-graphene oxide (MGO and MRGO) nanocomposites were made by Tanwar *et al.* [29] using graphite as a precursor material for the Hummers method of graphene formation and a simple synthesis method involving room-temperature coprecipitation and reduction processes. The energy transition levels in GO are clearly shifted as a result of the doping of magnetite, according to an analysis of the UV-visible and fluorescence spectra. Magnetite graphene oxide nanocomposites are naturally superparamagnetic, according to research on their magnetic properties conducted at room temperature using VSM. Environmental technologies make use of these graphene-based nanocomposites. By employing co-precipitation of Fe^{2+} and Fe^{3+} composited with graphene oxide (GO) in alkaline conditions, Li *et al.* [30] created Magnetic GO/Fe_3O_4. In this work, heterogeneous phases with a high concentration of oxygen-containing groups and a large specific surface area are observed in GO/Fe_3O_4. The characteristics of the GO/Fe_3O_4 surface were evaluated by means of SEM, XPS, FTIR, N_2 adsorption, and VSM techniques after the removal of malachite green (MG). The main adsorption-influencing parameters, including the GO/Fe_3O_4 dosage, pH level, and GO mass ratio, were investigated. Its stability, reversibility, and recoverability even allow for synergistic effects with plants, animals, or microorganisms in ecological remediation. Using a simple in situ co-precipitation method, Pu *et al.* [31] created a superparamagnetic graphene oxide (GO)/Fe_3O_4, which holds potential for use in wastewater treatment. The results showed that the synthetic MGO had a lot of potential for use as a practical adsorbent in wastewater treatment to remove organic contaminants. Co-precipitation produces MGO, but the process is simple, safe, affordable, controllable, and able to yield substantial amounts of material. Usually, two or three intricate reaction processes are needed for this process. Fe_3O_4 and reduced graphene nanoparticles may readily aggregate due to the potential disadvantages of the synthetic compound, and Fe_3O_4 may separate from graphene nanoparticles. Furthermore, this technique creates precipitates of the metal precursor, such as KOH or NaOH, using an alkaline precipitating solution. The degree of dispersibility decreases as a result of the reduction of functional groups added to GO [32].

2.2 Hydrothermal method

The solvent in the hydrothermal method is water. The metal salt solution is mixed with a predetermined percentage of alkaline solution, and the pH is then adjusted. The mixture is then allowed to crystallize in a high-pressure, high-temperature reactor. Magnetic nanomaterials can be created using heat treatment and separation [33, 34]. Teflon-lined stainless steel autoclaves with high temperatures and pressures are commonly used for the hydrothermal processing of MGO. This process not only reduces graphene oxide but also yields Fe_3O_4 nanocomposites and is economical, scalable, efficient, and environmentally benign. A one-step hydrothermal process was used by Ren *et al.* [35] to create superparamagnetic G–Fe_3O_4 nanocomposites. It is simple to control the size of Fe_3O_4 nanocrystals and their clusters by altering the initial mixed solvent ratio and reaction time, respectively. The hydrothermal method has two main

benefits because the reaction takes place in a sealed, high-pressure container: first, it can prevent component volatilization; second, by using high temperatures, it can improve magnetism [36].

2.3 Solvothermal method

For the high boiling point reducing agent, the solvothermal technique works better than the hydrothermal method because it uses organic solvents instead of water. In polar media, the synthetic compounds remained stable and had good dispersion throughout the solution process. Reaction conditions, such as temperature, time, reactant concentrations, solvent characteristics, and reactant ratios, can also be adjusted to alter the size, density, and crystallinity of Fe_3O_4 [37, 38]. Shen et al [39] used a simple and efficient one-pot solvothermal method to synthesize graphene-based magnetic nanocomposites (graphene/Fe_3O_4 and graphene oxide/Fe_3O_4). Fe_3O_4 nanoparticles, with a size of 60 nm, were definitely attached to the graphene sheets, preventing stacking. Wang *et al.* [40] synthesized a superparamagnetic graphene–Fe_3O_4 nanocomposite (G/Fe_3O_4) using a straightforward one-pot solvothermal technique. This nanocomposite was then used as an effective adsorbent to remove the organic. Fe_3O_4 spheres with a size range of 100 to 560 nm and uniform Fe_3O_4 nanoparticles with a size range of 2 to 100 nm can both be prepared using this technique. Despite being more expensive, the products made by the solvent thermal method have advantages over the hydrothermal method in terms of good dispersibility, high magnetism, and controllable morphology [41].

2.4 In situ chemical technique

Fe_3O_4/rGO is created by depositing Fe_3O_4 nanoparticles in situ on reduced graphene oxide (rGO) sheets using a straightforward, economical, effective, and environmentally friendly method as reported by Lim *et al.* [42]. The possibility of contamination in the final Fe_3O_4/rGO composite is eliminated in a single step of synthesis. Moreover, the one-step process can stop the aggregation of reduced graphene sheets. During the redox reaction, graphene oxide (GO) is reduced to rGO and iron(II) is increased in oxidation state to iron(III) [43]. Moreover, unlike other documented methods that call for high temperatures, like hydrothermal and chemical co-precipitation techniques, the Fe_3O_4/rGO nanocomposite forms at room temperature. As a result, the reported process is more economical and environmentally friendly than the alternatives due to its lower energy consumption [44].

2.5 Covalent bonding method

Diagboya *et al.* [45] developed iron oxide magnetic by using a covalent bonding method. The nanoparticles were bonded to graphene oxide (GO) at a temperature that was noticeably low using 3-aminopropyltriethoxysilane. Magnetic GO formed in-situ by magnetic structure formation on graphene/GO sheets, as well as magnetic GO synthesized by simply mixing graphene/GO with magnetic materials, are not completely stable. The instabilities are caused by weak interactions between magnetic materials and graphene/GO. Conversely, graphene/GO sheets are more stable and difficult to separate from the conjugated magnetic nanoparticles if a strong binding forms between them and the nanoparticles [46]. Fe_3O_4 NPs with surface modifications could be covalently bonded to GO for the first time. With the use of this technology, new graphene/GO–inorganic NP hybrids can be synthesized. Its primary benefits are that it can firmly graft Fe_3O_4 on the graphene surface through covalent bonding and predefine the loading amount of Fe_3O_4 [47].

2.6 Electrochemical method

Electrons must cross the electrode/solution interface and move from the electrode to the chemical compounds in the solution or the other way around in order to change the chemical composition of the mixture. This procedure is called electrolysis or electrochemical synthesis. Direct current (DC) from a constant source was utilized in the electrochemical reaction. Finally, a one-step process for creating superior graphene sheets is offered by electrochemical exfoliation of graphite [48]. Xie et al [49] prepared the precursor solution and cathodic deposition conditions appropriately to employ electrochemical synthesis to produce rGO/Cu composite films in a single step. Even when simple and environmentally friendly

electrochemical techniques are applied, there is little bonding interaction between Fe_3O_4 and graphene. The synthesis process is also gentle, rapid, and does not involve the needless use of hazardous reagents [50, 51]. Furthermore, the reaction is easily controlled and does not significantly alter the properties of the precursors. The ability to change the feeding rate is also advantageous [52, 53].

2.7 The ball mill method

One popular synthesising method for creating nanoparticles is mechanical milling, also known as mechanical alloying or mechanochemistry. High-energy milling is a popular technique for producing powder materials with sizes of a few tens of nanometers [54]. The ball mill method has the advantage of allowing MGO nanocomposites to be prepared without affecting the chemical structure of the GO framework. Obaidat *et al.* [55] used a mechanochemical method to ball mill graphene oxide and iron powder to create a large-scale GO-Fe_3O_4 nanocomposite. Furthermore, the GO functional groups may still be present in the ball-milled magnetic composites [56].

3. Sensors

3.1 Biosensors

Analytical markers known as biosensors exhibit hypersensitivity to extremely low dosages of the substance being studied and generate a measurable physicochemical signal. Graphene and its derivatives have demonstrated superior electrical conductivity when measured against other carbon-based materials. These features make them an excellent choice for electrochemical sensors. Important components like signal amplification, stability, and sensitivity are needed in electrochemical biosensing schemes to improve sensor performance (Figure 1). Due to the special characteristics of magnetic materials in combination with graphene and the synergistic effects of the two components, such as improved electronic conductivity and stability for biomolecules and large surface area for molecules immobilization, the creation of electrochemical sensors that sense metal ions, amino acids, H_2O_2, glucose, DNA, proteins, viruses, and other substances has received increased attention [57-59].

Depression, Parkinson's disease, tourette's syndrome, and other diseases and conditions have all been related to abnormalities in the body's dopamine concentration. GO/Fe_3O_4 was used by Siburianc *et al.* [26] to identify dopamine. When dopamine has a high peak current in both cathodic and anodic reactions, GO/Fe_3O_4 can detect it. To demonstrate this, the effectiveness of medication detection on a glassy carbon electrode with and without modification was compared. Ascorbic acid, glucose, and uric acid were tested as interfering agents for the GO/Fe_3O_4 sensor; GO/Fe_3O_4 exhibits good selectivity. After three weeks, current peaks dropped to 66% performance under storing durations.

Baghayeria *et al.* [60] made use a simple incipient wetness impregnation method to functionalize MGO with amine-terminated poly(amidoamine) dendrimer and set it up with palladium nanoparticles (GO-Fe_3O_4-PAMAM-Pd). The surface of the magnetic GO composite is notable for having terminal amine groups, which can provide a large number of Pd nanoparticle-friendly binding sites. Furthermore, the special hyperbranched structure of PAMAM allows for better dispersion of palladium nanoparticles, increasing the electrocatalytic activity of the modified electrode. The proposed H_2O_2 sensor had an excellent reproducibility (R.S.D. = 2.1%) and a low detection limit of 0.01 M. A successful study was also conducted to determine whether the GO-Fe_3O_4-PAMAM-Pd/GCE could be used to measure H_2O_2 in real water samples.

Souza *et al.* [61] produced a novel structure entirely covered in manganese ferrite nanoparticles that resembles a crumpled ball of graphene in a single step. The suggested H_2O_2 sensor was the most sensitive and had the lowest detection limit.

Meanwhile, it is vital to identify and track several small molecules, such as amino acids, hormones, H_2O_2, glucose, and more, in order to unearth certain biological or clinical processes. To tackle this, Jiang *et al.* [62] synthesized Fe_3O_4 nanoparticles on GO/PG in situ to produce a novel Fe_3O_4/GO/PG ternary composite. They subsequently constructed an electrochemical sensor that can successfully detect DA and H_2O_2 using this composite. Comparing

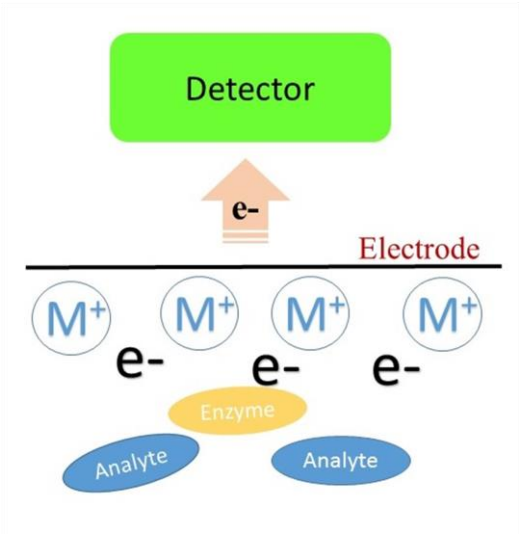

Figure 1. schematic configuration of an electrochemical biosensor [63].

Fe_3O_4/GO/PG0.88 to bare GCE and Fe_3O_4/GO reveals remarkable properties. These characteristics are connected to strong electrical conductivity of GO/PG, its excellent water solubility, and catalytic activity of Fe_3O_4. The sensor based on Fe_3O_4/GO/PG0.88 exhibits good DA and H_2O_2 sensitivity. H_2O_2 and DA have linear detection limits as well as ranges of 0.30-30 M and 0.50-277 M, respectively. K. Nayak *et al.* [64] produced magnetic nanocomposites of graphene oxide, which were utilized as an electrode material for bio receptor loading. The improved surface area with high electric conductance enhanced the sensor response. Progesterone (PGN) antibody immobilization on the altered electrode-sensing surface prevented electron transport, which in turn decreased the current response. Under ideal experimental conditions, the measured response reduced linearly with increased PGN concentration range of 0.01 pM-1000 nM, with good detection limits of 0.15 pM (DPV) and 0.17 pM (CV). The label-free electrochemical immunosensor exhibits high stability, repeatability, sensitivity, and selectivity in water samples, making it a promising platform for quick and easy PGN analysis.

Reduced MGO may offer immobilized biomolecules an easily controlled, biocompatible microenvironment through the use of a magnetic field. Poo-arporn *et al.* [65] developed a novel type of disposable electrochemical sensor for the rapid measurement of ractopamine using the Fe_3O_4/rGO nanocomposite. The results showed that the sensor had a low detection limit, good stability, sensitivity, repeatability, and acceptable selectivity. It was also easy to prepare. It also had good electrochemical properties. The proposed sensor could be used to measure the amount of ractopamine in the real pork samples if it recovers well.

Magnetic reduced graphene oxide, developed by Liu *et al.* [66] can be used in conjunction with a screen-printed electrode and a portable potentiostat to detect, quantify, and capture miRNA 183-5p. MicroRNA (miRNA) is a type of noncoding RNA that regulates cellular functions and gene expression. Many oncogenic miRNAs are expressed abnormally in prostate cancer, implying that they could be used as biomarkers. When combined with a disposable printed electrode and a portable potentiostat, they developed multifunctional nanosheets capable of capturing, detecting, and quantifying miRNA 183-5p from prostate cancer cells. MrGO, screen-printed carbon electrodes, and portable potentiostats can all be used to accelerate biomarker identification, simplify the procedure, and reduce the cost and length of analysis.

The methylation status of the O6-methylguanine-DNA methyltransferase (MGMT) gene is critical for the diagnosis, prognosis, and management of glioblastoma. Immunological recognition-based electrochemical method is now a simple and accurate way to detect MGMT gene methylation. Wang et al. [67] suggested the use of an electrochemical immunosensor to detect MGMT gene methylation. The electrode was coated with a graphene oxide-magnetic nanoparticles-β-cyclodextrin nanocomposite, and the host-guest interaction between the β-CD and anti-5-methylcytosine (5mC) antibody immobilized the antibody on the improved electrode. The immobilized antibody was able to bind to the target directly without the need to create specific sequences. Redox recycling-assisted signal amplification increased the sensitivity of the developed immunosensor, and a wide linear range (0.001-1000 nM) was obtained for the methylation evaluation of the MGMT gene, with a detection limit of 0.0825 pM.

Rutin (3',4',5,7-tetrahydroxyflavone-3-d-rutinoside) is a common flavonoid glycoside that is current in a wide variety of plants, particularly buckwheat plants. According to recent pharmacological research, rutin inhibits hemostasis, vascular rupture, and capillary wall permeability and fragility. Moreover, it possesses analgesic, antibacterial, anti-inflammatory, anti-radiation, anti-oxidant, and anti-myocardial hypoxic qualities. Moreover, it lessens platelet aggregation, diuresis, spasmolysis, and serum cholesterol. Thus, it is essential to create straightforward, dependable, and sensitive techniques for rutin analysis. Deng *et al.* [68] created an electrochemical sensor that uses electrochemically reduced graphene (NH_2-Fe_3O_4 NPs-ErGO) as the sensing material and a nanocomposite of amine-functionalized Fe_3O_4 nanoparticles for the detection of rutin. They discovered that the GCE modified by nanocomposites (NH_2-Fe_3O_4 NPs-ErGO/GCE) greatly increases the rutin oxidation signals. Ascorbic acid interferences have also been successfully eliminated using this modified electrode. The detection limit (S/N = 3) was 4.0 nM. The developed method for rutin determination in pharmaceutical tablets and urine samples is sensitive and selective. Along with its many apparent benefits, this innovative method offers low cost, high sensitivity, fast response, and simplicity.

Since high concentrations of the protein can be cytotoxic, the development of new control technologies hinges on the identification of a novel method for calculating patulin concentrations. In order to quantify the amount of patulin toxin using the square wave voltammetry technique, Using a composite of magnetic nanoparticles/Fe_3O_4/GO and ionic liquid-based molecularly imprinted polymer (MIP), Mohadesi *et al.* [69] have created a novel sensor. The increased selectivity of sensor was boosted by the special patulin molecular cavities in MIP, while its increased sensitivity was ascribed to the superior conductivity of GO nanosheets. In actual apple juice samples, the patulin toxin was successfully identified using the MIP/Fe_3O_4/GO/GCE method.

3.1.1 Biosensor mechanism

An electrode based on GOx was used to measure glucose, as reported in a report [70] described the method for measuring glucose using an electrode based on GOx. Based on the reduction in the reduction current, the glucose concentration can thus be ascertained and employed as a biosensor system for reagentless glucose sensing. The following is an explanation of the electrocatalysis mechanism:

1. $GOx(FAD) + 2H + +2e - \rightarrow GOx(FADH_2)$

2. $GOx(FADH_2) + O_2 \rightarrow GOx(FAD) + H_2O_2$

This process forms FAD by combining oxygen and FADH2. Equation 3 shows how the reaction of glucose inhibits an electrocatalytic reaction in Equation 1 when glucose is added to an air-saturated system. Consequently, FAD lowers.

3. $GOx(FAD)$ + glucose $\rightarrow$ $GOx(FADH_2)$ + gluconolactone

The square wave voltammogram data demonstrate that the reduction current caused by the inhibition of FAD electrocatalytic activity decreases with increasing glucose concentration. Consequently, the operational potential of the glucose biosensor is restricted. Because the Fe_3O_4 and rGO sheets complement each other well to form a biocompatible carbon environment with good conductivity, the rGO/Fe3O4/GOx/GCE exhibits good electrocatalytic activity [71].

Fe_3O_4-GBR was used in Ray *et al.*'s cholesterol sensing mechanism (Figure 2) [72]. When cholesterol and cholesterol oxidase react, a specific amount of hydrogen peroxide is released. The kinetics study demonstrated that Fe_3O_4-GBR has a high binding affinity for both H_2O_2 and TMB. This implies that Fe_3O_4-GBR may be able to cling to more H_2O_2 molecules due to its small size and large surface area. The labile C-Br bond in GBR makes it easier for ·OH radical species to form from H_2O_2. It is possible that H_2O_2 molecules adsorbed on the Fe_3O_4 surface will also activate, leading to the production of ·OH radicals, since Fe^{2+} and Fe^{3+} iron ions have the ability to escape from Fe_3O_4 nanoparticles at low pH values. Fe^{2+} may become Fe^{3+} as a result, and the resulting ·OH radicals may oxidize TMB to oxyTMB. Fe_3O_4-GBR can produce ·OH radicals when Fe^{2+} oxidation and dissociation of the labile C-Br occur simultaneously because it contains both Fe_3O_4 and GBR. Thus, single-electron transfer will occur in the produced ·OH radicals to aid in TMB oxidation, resulting in a color shift from colorless to blue. Furthermore, lower

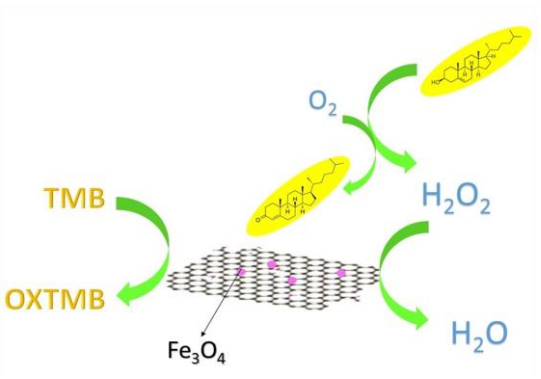

Figure 1. Mechanism of cholesterol sensing [72].

(acidic) pH values typically speed up the formation of ·OH radicals. Therefore, a key factor in the oxidation of TMB into oxyTMB is the quicker generation of ·OH radicals in the presence of Fe_3O_4-GBR.

The electrochemical cyclic voltammetry graph is used to demonstrate the conversion of H_2O_2 to $H_2O + O_2$ in the micrographic images provided by Sobahi *et al.* in Figure 3. The graph in [73] shows how the H_2O_2 detection slope changes over time. The reduction/oxidation process at the electrode surfaces (Fe_3O_4/Gr/CC electrode) is depicted in Figure 3 to aid in the detection of H_2O_2. Fe_3O_4/Gr NC reduces H_2O_2 via electron transfer to produce hydroxyl ions (OH) and, when combined, water and oxygen molecules.

Fe_3O_4/GO magnetic nanocomposite was employed by Rodthongkum *et al.* [74] to separate CBF and CBZ; Fe_3O_4/GO was added to the mixed pesticide solution. Fe_3O_4 nanoparticles have proven useful for selective pesticide extraction due to their apparent magnetism and ability to separate faces using an external magnetic field as a sensing mechanism. Although π-π plays a major role in the interaction of GO with pesticides, self-preconcentration results from enhanced analyte adsorption to the Fe_3O_4/GO nanocomposite surface due to the interaction of GO with the aromatic ring of pesticides. Therefore, without requiring sample preparation, the synergistic interaction between Fe_3O_4 and GO, along with their individual characteristics, allows for the selective detection of CBZ and CBF. Using a magnet, the binding between Fe_3O_4/GO and the pesticides was extracted from the mixture.

3.2 Gas sensor

A high-performance gas sensor that can identify NO_2 at room temperature was created by Zou *et al.* Reduced graphene oxide and γ-Fe_2O_3 are combined in a 3D core-shell film, which was applied with the aid of a magnetic field during the drop-coating process [75] The sensor is superior to a 2D RGO sensor because of its good selectivity and high sensitivity. Even at a low NO_2 concentration of 100 ppb, it retains a response amount of 1.23. The consistency of the sensor is increased by fabricating it in a magnetic field. Using the findings of this study, large-scale and useful gas sensors can be produced [76].

Using density functional theory calculations, Impeng *et al.* examined the viability of employing MnN4-GP (MnN4 moiety embedded graphene) as a gas sensor. Thirteen gas molecules were studied for their

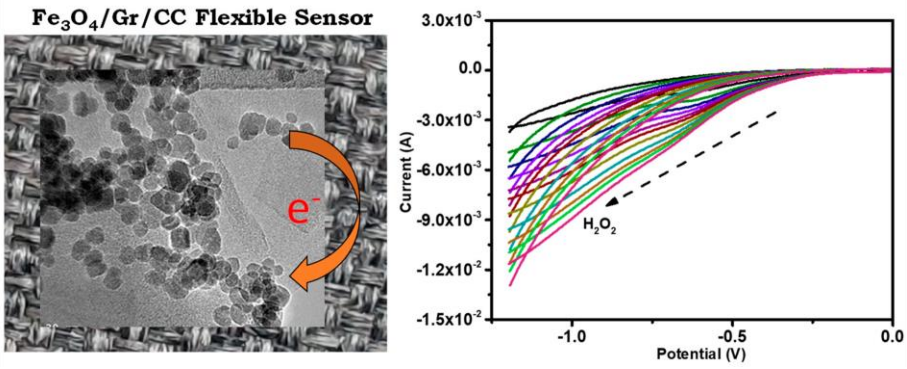

Figure 3. Schematic for the mechanism towards the detection of H_2O_2 on Fe_3O_4/Gr/CC [73].

adsorption behavior, electronic characteristics, and magnetic properties on MnN4-GP. It was discovered that whereas the other gases displayed weak physisorption, CO, NO, NO_2, SO_2, and O_2 demonstrated strong adsorption. NO had the highest adsorption energy of all the gases. Only electrical and magnetic characteristics of the CO, NO, and NO_2 chemisorptions were altered. Following the adsorption of NO_2, NO, and, CO the magnetic moment of MnN4-GP decreased. We measured the rates of recovery for NO, CO, and NO_2; CO recovered the quickest. Taking into consideration gas interactions, charge transfer, magnetic moment, density of states, and recovery time, a highly selective and sensitive gas sensor for CO detection with a short recovery time is proposed in MnN4-GP [77, 78].

rGO–$CuFe_2O_4$ nanocomposite was created in the study by Achary *et al.* through a low-cost, one-step combustion process. Utilizing the superior electrical qualities of rGO in conjunction with the sensing properties of $CuFe_2O_4$, this nanocomposite was employed as a high-performance NH_3 gas sensor. The study demonstrated for the first time how highly selective and sensitive the rGO-$CuFe_2O_4$ sensor is for NH_3 gas detection at room temperature. The combination of rGO and $CuFe_2O_4$ enhanced the sensing ability of $CuFe_2O_4$. In terms of sensitivity at room temperature, the sensor performed better than other gases, responding 25% for 200 ppm and 2% for 5 ppm of NH_3. Furthermore, the sensor showed response times of three and six seconds, respectively [79, 80].

Qin *et al.* [81] looked into the sensitivity of graphene combined with semiconductor metal oxides like γ-Fe_2O_3 and SnO_2. Following the creation of the nanoparticles, they studied how sensitive they were to various gases, including formaldehyde, methanol, ethanol, isopropanol, H_2S, CO, and NO. At 100°C, the γ-Fe_2O_3/SnO_2/RGO composite was sensitive to organic vapors; however, as the temperature increased, its sensitivity to H_2S decreased. Reaction times did, however, increase with temperature; at 200°C, H_2S demonstrated an astounding 6-second response time. The results offer insightful information for future uses of gas sensing technologies [82].

Tung *et al.* used reduced graphene oxide sheets and immobilized magnetic nanoparticles to create a gas and vapor sensing platform [83]. They created five, ten, and twenty nm-sized nanoparticles and investigated how they affected sense capacities. In contrast to bare reduced graphene oxide, the 20 nm particles exhibited greater sensitivity to gases at 1–5 ppm concentrations than the 5 and 10 nm particles did. In addition to being incredibly sensitive to trace amounts of ethanol vapor (1 ppm) and other volatile organic compounds (VOCs), the sensors were highly selective for NO_2 gas [84].

Using a homotaxial electrospinning method, Guo *et al.* successfully synthesized reduced GO)/α-Fe_2O_3 composite nanofibers and pure α-Fe_2O_3 nanobelts [85]. At 375°C, gas sensors that employed 1% rGO/α-Fe_2O_3 nanocomposites demonstrated an enhanced ability to respond to acetone, reaching a maximum sensitivity of 8.9 for 100 parts per million of the acetone gas. In addition, the sensors demonstrated quicker recovery and response times—3 and 9 seconds, respectively—than pure α-Fe_2O_3 NBs. Ohmic contact formation, rGO nanosheet imperfections, and rGO catalytic activity in adsorption, dissociation, and transportation are all responsible for this improvement. Furthermore, because of its unique two-dimensional π–π conjugated structure and the electrons it transfers to α-Fe_2O_3, rGO in combination with that oxide enhanced the gas response and reduced the resistance of the sensor [86].

A straightforward and low-cost technique for creating graphene oxide (GO) gas sensing devices embellished with strontium titanate perovskite ($SrTiO_3$) is presented by Kacem *et al.* The study looks into the gas sensing capabilities of graphene oxide (GO) that has been decorated with $SrTiO_3$ as well as plain GO. The findings show that, in comparison to pristine GO, GO/$SrTiO_3$ sensors have better sensing abilities. GO/$SrTiO_3$ is the most responsive and sensitive to NO_2 compared to NH_3 or CO_2. Furthermore, GO/$SrTiO_3$ sensors respond consistently to NO_2 even at varying humidity levels, and they show a lower limit of detection of 72 ppb for NO_2, which is higher than that of pure GO [87, 88].

Ma *et al.* report the alignment of three-dimensional Fe_3O_4@SiO_2@rGO core-shell spheres caused by a magnetic field, which results in multichannel gas sensors. By adjusting the magnetic field and sphere concentration, these sphere-based sensing channels can be customized. Results exhibit that the

multichannel 3D Fe_3O_4@SiO_2@rGO sensor has a high selectivity towards 5 ppm of NO_2 at room temperature, good response stability, and an ultrahigh sensitivity of 34.41. This sensitivity is approximately 7.96 times higher than a random 3D rGO gas sensor. According to the study, the multichannel sensing devices have better humidity resistance, response repeatability, selectivity, and sensitivity to NO_2 gas. The enhanced sensing performance is attributed to the efficient use of the large sensing area offered by the rGO nanosheets. Moreover, other nanomaterials loaded onto the surface of magnetic spheres can have their sensing capabilities improved [89, 90].

Prussian blue (PB) was used by Ruixue Mo *et al.* to successfully create porous Fe_2O_3 nanocubes by acting as a metal-organic framework (MOF) and self-template. Using a solution method and calcination process, porous Fe_2O_3 nanocubes and reduced graphene oxide (rGO) were combined. The microstructure, element composition, and morphology of the resultant nanocomposites were examined. When compared to pure Fe_2O_3 nanocubes at room temperature, the gas sensor based on the porous Fe_2O_3 nanocubes combined with rGO demonstrated improved gas sensing performance for n-butanol. The enhancement ranged from 12.7% to 171% for 100 ppm n-butanol. The gas sensor also demonstrated excellent linearity, long-term stability, and selectivity. Because of its high surface area porous morphology and the formation of a p-n heterojunction between rGO and the porous Fe2O3 nanocubes, the gas sensor exhibits improved stability, selectivity, and response. [91, 92].

Munusami *et al.* used a hybrid structure of $ZnGa_2O_4$ and graphene (ZGO/GR), synthesized by a hydrothermal process, to develop a high-performance gas sensor for liquid petroleum gas (LPG). It was looked into whether the ZGO/GR sensor could identify a range of gases at various ppm concentrations. When compared to other gases, the results demonstrated exceptional sensing performance, particularly towards LPG gas. Good selectivity, long-term stability for LPG gas, high sensitivity, quick response, and quick recovery were all demonstrated by the hybrid ZGO/GR sensor. The synergistic effects between $ZnGa_2O_4$ and graphene, which increase adsorption/desorption capacity of gas molecules, facilitate carrier transfer, and form local heterojunctions between $ZnGa_2O_4$ and graphene, are what lead to the enhanced sensing response of the ZGO/GR composite sensor [93, 94].

3.2.1 Gas sensing mechanism

In the gas sensing mechanism of graphene and its derivatives, target gas species temporarily substitute dopants, altering the localized electronic concentration of the graphene layer. The gas sensing material is exposed to holes (H_2O and NO_2) or electrons (NH_3 and CO) as a result of this doping effect [95, 96].

The oxygen functional groups in graphene oxide (GO) act as active sites for gas adsorption, facilitating charge transfer between GO and the target gas. The low conductivity of GO, however, limits its sensitivity. The performance of gas sensing is enhanced when $SrTiO_3$ is added to GO layers. This enhancement results from the interaction between $SrTiO_3$ and GO, which is highly reactive. The dominant carriers in the material and the gas species (oxidizing or reducing) determine whether exposure to a target gas raises or lowers sensor resistance. Although SrTiO3 is an n-type semiconductor, because GO predominates, the GO/$SrTiO_3$ nanocomposite exhibits p-type behavior. When molecules come into contact with an oxidizing gas like NO_2, the oxygen functional groups in GO help them adsorb NO_2, lowering resistance. As electrons are transferred via the n-type $SrTiO_3$ to the p-type GO, resistance increases. The interaction of the active layer with atmospheric oxygen traps electrons, which leads to the formation of a depletion layer that accumulates positive carriers. More electrons are trapped when exposed to NO_2, and the GO-supported n-type $SrTiO_3$ nanoparticles offer more sites for NO_2 adsorption. As the electrical charge of GO is transferred to $SrTiO_3$ nanoparticles, resistance is consequently greatly decreased. This explains the greater response of GO/$SrTiO_3$ nanomaterials to NO_2 compared to bare GO [87, 97].

γ-Fe_2O_3@RGO (graphene oxide) uses resistance changes driven by oxygen chemisorption on the surface of material and target gas (NO_2) reactions as its gas sensing mechanism [98]. The mechanism can be summed up as follows:

1. When exposed to air, oxygen molecules cling to the surface of RGO, stealing electrons. As a result, on the

surface of RGO, chemisorbed oxygen species (O^-, O^-_2) and an electron depletion layer (EDL) form.

2. When NO_2 gas is present, NO_2 molecules enter the RGO electron-depleting layer (EDL) and mix with the oxygen species that has already been adsorbed (O^-_2) to form NO_2^-. This response takes place as follows:

$O_2(air) \rightarrow O_2(ads)$ (1)

$O_2(ads) + e^- \rightarrow O^-_2(ads)$ (2)

$NO_2(gas) + e^- \rightarrow NO^-_2(ads)$ (3)

$NO_2(gas) + O^-_2(ads) + 2e^- \rightarrow NO^-_2(ads) + 2O^-(ads)$ (4)

1. Differenciates: When RGO and γ-Fe_2O_3 combine to form heterostructures, conductivity is increased and more active sites for NO_2 adsorption are available. Electron mobility is enhanced when electrons are transferred from RGO to γ-Fe_2O_3, balancing the Fermi level and forming an ohmic contact. As a result, there are more chemisorbed oxygen species produced on the γ-Fe_2O_3 surface, which raises the quantity of active sites for hole formation and the NO_2 reaction.

2. The 3D Shell-Core Architecture: The three-dimensional core-shell structure of γ-Fe_2O_3@RGO prevents RGO from clumping together while improving NO_2 gas molecule diffusion and transport. There are more channels open for gas diffusion as a result of the separated RGO layers and the avoidance of RGO sheet stacking.

3. Greater Surface Area: The larger surface area of the 3D γ-Fe_2O_3@RGO hybrids gives more areas for NO_2 gas adsorption, which further enhances the performance of the sensor.

When combined, these elements improve the sensing performance of the 3D γ-Fe_2O_3@RGO sensor [76, 99, 100].

4. Conclusion and future perspectives

This review went into great detail about the evolution of MGOs and other sensor types. Many MGOs have been synthesized successfully using covalent and non-covalent binding of graphene and magnetic nanoparticles, as well as in situ synthesis of magnetic nanoparticles on graphene surfaces. Magnetic graphene materials have a large surface area, excellent absorption properties, biocompatibility, water solubility, electron transfer, and high saturation magnetization. These characteristics make them ideal for environmental and biomedical applications. Magnetic graphene materials are simple to synthesize and functionalize to provide the precise properties required for a variety of applications. Researchers discovered a wide range of biosensors and gas sensors based on these composites and their remarkable properties; these sensors demonstrated enhanced selectivity and sensitivity, as well as a combination of properties.

The synergistic benefits of MGOs have increased the use of sensors in a variety of fields. MGOs, on the other hand, are still in their infancy, and numerous issues must be addressed. The bottlenecks are currently MGO preparations with controlled sizes and shapes, as well as low-cost, high-yield methods. The potential applications of MGO in other fields, such as medication delivery, catalysis, and energy storage, need to be investigated further. New synthesis techniques, as well as the development of improved ones, will be required to improve the properties of MGO and broaden its applications. When all factors are taken into account, MGO has a lot of future potential for use in a variety of industries, as well as the advancement of sophisticated sensing technologies.

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