

REVIEW ARTICLE

Graphene Oxide and Based Materials: Synthesis, Properties, and Applications – A Comprehensive Review

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ABSTRACT: Graphene-based materials have garnered significant interest in the burgeoning field of contemporary flexible and bendable technologies. Graphene has emerged as a prime candidate for the development of flexible electronics due to its outstanding electrical, mechanical, and optical properties, coupled with the ease of functionalizing its derivatives. This review provides an exhaustive analysis of the latest advancements in the synthesis and applications of graphene-based composites. Exceptional properties of Graphene including high thermal conductivity, chemical stability, optical transparency, high current density, and increased surface area, have broad implications across thermodynamics, chemistry, optics, and mechanics. The growing use of graphene oxide materials across various industries has fueled a surge in interest. The synthesis of graphene oxide compounds follows established procedures such as those by Brodie, Staudenmaier, and Tour, with ongoing discussions on large-scale production using various oxidants. Recent developments have led to numerous new, enhanced, and modified Hummers techniques for producing graphene oxide. Graphene oxide (GO) serves as a crucial foundational material for a wide range of applications, including energy storage, water purification, and as a composite in biosensors, electronics, and hazardous gas removal. This review delves into these applications, highlighting GO's role in improving the efficiency and performance of these technologies. Additionally, the paper addresses the current challenges and obstacles in the development and application of graphene, providing insights into potential solutions and future research directions. By offering a comprehensive and practical database, this review aims to facilitate the future development of graphene-based composite materials, driving innovation and advancements in various high-impact applications.

Keywords: Graphene oxide, Synthesis methods, Properties, Multifaceted applications, Energy storage devices, Photocatalysts.

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1. INTRODUCTION

Since new materials are frequently required in order to realize new technologies, it is widely accepted that materials play a crucial role in the evolution of science and technology [1]. As a result, discovering better materials has long been a prominent field of scientific inquiry. Graphene is the "miracle material" with the highest electrical and thermal conductivity on the planet [2]. It is the world's thinnest, toughest, strongest, and most potent substance. It is a crucial part of other significant allotropes [3]. A type of graphite that can be treated physically and oxidative is called GO. In response to environmental concerns and demand for materials, there has

been great interest in both academic research and commercial applications of graphene and its oxidation states. Its distinctive and intriguing mechanical, electrical, and other features are to blame for this [4]. Along with carbonyl (C = O), hydroxyl (-OH), alkoxy (C-O-C), carboxylic acid (-COOH), and other functional groups, GO also contains oxygen functional groups. Graphene and this carbon's hexagonal structure are comparable [5]. In terms of cycle stability, GO beats out graphene owing to its greater performance and lower price, graphene oxide is preferred to graphene as an electrode material for supercapacitors [6,7].

There are currently more opportunities and problems in numerous diverse scientific domains related to the synthesis and characterization of graphene (G) and GO. Researchers are currently focusing on updating procedures for making GO from waste graphite (rGO) and other natural resources [8]. The general synthesis process of G has been studied in the early stage, but there are not many reports on innovative GO synthesis technology [9].

The method of making graphene oxide was first developed in 1859 [10]. At that time, the product of the Brody reaction between graphite and KClO_3 in fuming nitric acid contained more flake graphite [10]. Staudenmaier later improved on the early method in 1898 by adding concentrated H_2SO_4 to the mixture to make it more acidic, and by adding chlorate in a series of aliquots during the reaction [11]. Hummer and Offeman subsequently improved the process using alternating oxidation. Graphite undergoes an oxidation process that includes concentrated H_2SO_4 , NaNO_3 , and KMnO_4 . This method is currently used most of the time [12] with only minor improvements and adjustments.

The production process of graphene is currently being studied in a variety of ways [13]. Using micromechanical exfoliation technique, Novoselov and Heim were the first to identify single-layer graphene in strongly oriented pyrolytic graphite [14]. Although graphene is easily prepared, the extremely low yield makes it hard to carry out high-quality industrial manufacturing [15]. The uses chemical vapor deposition (CVD) technology to produce "petal" graphite sheets that are 20 nanometers thick or less. Although the CVD process produces graphite sheets that are much thinner than before, adding additional nickel impurities is a completely new problem [16]. Solvent stripping method is a recent and cutting-edge strategy [17].

By functionalizing GO, its electrical properties can be significantly altered [18]. The resulting chemically modified graphene (CMG) may have many more uses, with almost unlimited uses [19]. GO may be functionalized in a variety of ways depending on the desired use [20].

The most typical technique for mass production of chemicals based on graphene is chemical synthesis [21]. However, well-known chemical synthesis processes, including the Brodie, Staudenmaier, and Hummer processes, are toxic and hazardous due to the release of ClO_2 and NO_2 during the process [22].

The creation of hydrogen bonds between polar groups on the surface of GO and water molecules results in the development of stable GO colloidal suspensions, giving GO

an edge over other carbon-based materials in terms of possible biological uses [23]. Graphene has found widespread use in healthcare, biology, and environmental technologies [24]. Antimicrobial compounds are often utilized nowadays to safeguard public health. A helpful nanomaterial for killing bacteria is graphene oxide [25]. It is an excellent material for photocatalysis, electrochemical treatment, and the adsorption of dangerous compounds in wastewater treatment processes because of its enormous surface area, high current density, and high light transmittance [26]. In summary, making this substance is necessary to make graphene applications a reality [27]. The quality of the material is largely affected by GO, a by-product of the preparation process that is thought to offer important prospects for graphene synthesis [28]. Thus, it is essential to research the characteristics of graphite oxide [29].

Electronics, energy storage devices, (bio)sensors, biomedical applications, supercapacitors, membranes, catalysts, and water filtration all employ rGO (figure 1). rGO is used as a chemical and biological sensor in electrical devices known as field-effect transistors (FETs) [30].

GO damages cells via a variety of mechanisms, including reactive oxygen species (ROS), DNA deterioration, and plasma membrane rupture [31]. Most people think of rGO flakes as a special type of chemical graphene derivative [32]. Reduced graphene, chemically modified graphene, and chemically changed graphene are all names for rGO [33].

Cancer treatment, medication administration, and cell imaging are some of the current applications of GO, an oxidized derivative of graphene, in biotechnology and medicine [34]. Other non-metallic materials cannot match graphene's special characteristics. Due to some of its similarities to semiconducting metals, it is categorized as a semimetal [35].

The world's first car with a graphene body is on display in Manchester [35]. Briggs Automotive built the car in Speke, Liverpool, with panels made from graphene. The BAC Mono was the inspiration for an exhibition showcasing future technology graphene, which is 200 times stronger than steel. Biomedical devices, electronics, energy storage, sensors, coatings, and composites are just a few of the projects that use it [36]. It is the perfect material for applications requiring tissue engineering and medication delivery because of its enormous surface area and biocompatibility [37].

Long before graphene was really discovered, in 1859 Benjamin Brody produced graphene oxide for the first time [38]. He created a process for producing graphite oxide. It undoubtedly produces large amounts of single-layer graphene oxide, and requires oxidation and exfoliation of part of the naturally occurring crystalline [39].

GO is produced by exposing graphite to strong oxidants such as fuming nitric acid and potassium chlorate [40]. According to some studies, graphene oxide can serve as a nano-carrier for the anticancer drugs SN-3822, doxorubicin, camptothecin, and methotrexate. Drug loading efficiencies as high as 200% were also recorded for DOX/NGO25. The two-dimensional structure of GO, which shares the same atoms with human organs, tissues, and cells, can lead to odd

interactions with biological membranes and blood proteins that can have negative effects including the activation of immune cells and blood clots.

GO contributes to the spread of cancer by interacting with the plasma membrane and promoting TGF signaling-dependent epithelial-mesenchymal transition [42]. Graphene and graphene oxide differ in size and physicochemical characteristics, which affect how they interact with the skin [41]. According to various studies, there is little indication that skin cells are harmful when exposed to high concentrations of graphene and its derivatives for prolonged periods of time [43].

Packaging and electrochemical sensing in the food industry have seen the successful entry of graphene derivatives [44]. We have not yet fully analyzed all of their multiple uses in the fields of analysis, milk and beverage technology.

Strong oxidizing substances, such sulfuric acid and graphite, are used to create it artificially [45]. Scientists from all around the globe are interested in graphene because of its extraordinary features, which include a high surface area, mechanical stability, and adjustable electrical and optical properties [46]. Sensing, biology, nanoelectronics, and energy. Graphite derivatives are more beneficial than the parent substance [47].

In contrast to electric double layer capacitors (EDLCs), pseudocapacitors typically contain conducting polymers and metal oxides [48]. One of the most intriguing potentials is

graphene, which has a large surface area, great electrical conductivity, exceptional mechanical and electrochemical resilience, and exceptional flexibility [49]. Graphene/MHS nanocomposites feature improved mechanical strength, effective use of MHS materials, and higher electrical conductivity due to the high surface area and superior electrical conductivity of graphene sheets [50,51].

Therefore, it prevents structure collapse due to layer elongation and allows simple electron transport within the material structure, both of which are necessary for enhanced electrochemical performance [52,53]. This method also utilizes microwave irradiation and solvothermal effects to fabricate graphene nanosheets in 5-15 minutes at a temperature of approximately 300 °C, opening the door to simple and economical fabrication [54,55].

Experts and industry agree that Hummers' strategy is the most promising for GO synthesis [56]. Most of these modified versions are known as modifications or enhancements to Hummers' method [57]. Researchers often modify Hummers' synthesis methods to obtain desired properties and final applications. However, it has only been applied thus far to produce GO, the starting material for RGO and graphene. The current work is the first to create innovative graphene oxide-cellulose nanocomposites (GO-CNC) [58] in situ using a direct one-pot method. Cellulose for this experiment was created from waste jute fiber [59]. The ability of the developed nanocomposites to absorb the harmful azo-chromic methylene blue was investigated.

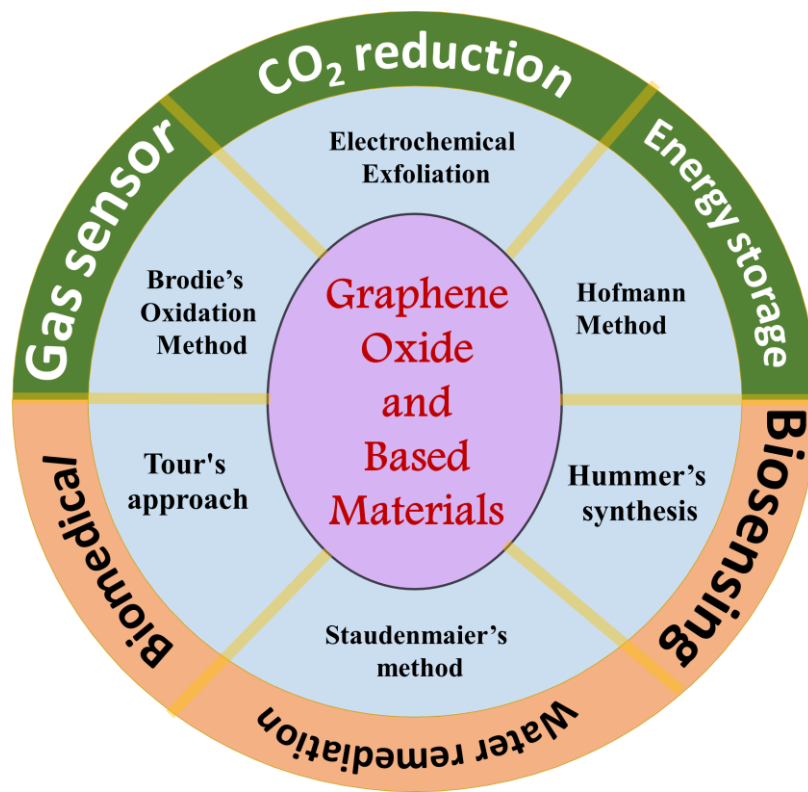


Fig. 1. Different methods for the synthesis of GO and Based materials for various application.

The adsorption experimental results were analyzed thermodynamically, isothermally, and process kinetically [60]. Use response surface technology to improve procedures and obtain maximum adsorption efficiency. This study is noteworthy because it raises awareness among the scientific community and the business sector of a novel in situ method for removing colors from wastewater utilizing ground-breaking GO-cellulose nanocomposites [61]. This novel biocomposite is of great interest due to its simple bioderivative process, biodegradability, low cost, low biocompatibility, low toxicity, [62] and affinity for methylene blue dye. It has significant promise for a variety of industrial uses in the real world [63].

Two-dimensional nanomaterial GO has a high adsorption capacity, strong mechanical strength, and excellent heat conductivity [64]. GO has been employed in the faster selective oxidation of alcohols and carbon monoxide as well as the manufacture of fuel and environmental purification. It may also be used for organic compound hydrogenation, nitroarenes reduction, and cross-coupling reactions [64].

The hunt for more resilient carbon catalysts has been successfully supported by the 3D network of GO, also known as a carbon monolith. Metals that are employed in several chemical applications and as catalysts are now coated on 2D and 3D GO [65]. For instance, Ru is a stable metal at low temperatures, but at high temperatures it changes into different ruthenium-based oxides (RuOx) and hydrated ruthenium oxides (RuO₂.xH₂O). On carbon substrates like graphene oxide, carbon nanotubes, and mesoporous carbon, Ru and Ru oxides [66] can be deposited to improve the overall conductivity and catalytic activity of these systems.

Gold nanoparticles have been adsorbed on less expensive inorganic and organic substrates in order to change the surface functionality and improve catalytic activity [66]. The findings demonstrate the amazing stability and application of the Au-supported GO catalyst for gas-solid oxidation processes. Last but not least, palladium-based catalysts have been shown to have improved oxidative activity for reactions involving alcohols, hydroxazine and the Suzuki reaction of unsaturated hydrocarbons. Palladium, a noble metal utilized as a catalyst, [67] has received a lot of interest due to its activity, accessibility, and stability. It is shown here that employing X-ray synthesis on supports with complicated 3D geometries, [68] well-dispersed nanoparticles of the aforementioned metals may be generated, which is not always attainable using normal chemical procedures. This capacity has the potential to speed the development of active catalysts for a variety of industrial applications [69]. Academic institutions are where most graphene research is done. There are several uses for these stacked two-dimensional (2D) materials [70]. Vertical alignment for 2D nanomaterial structures on carbon-based substrates allows for extremely efficient active sites and regulated ion transport, resulting in improved electrochemical performance [71].

Both mechanical exfoliation and hydrothermal redox techniques have been investigated as important synthetic

strategies to generate graphene. Techniques include electron bombardment, CVD, and top-down synthesis [72]. Functional graphene has been created using the aforementioned procedures in a variety of shapes and for a number of purposes. The energy and environmental industries have always paid close attention to research on graphene. These components can also be combined with other substances to create practical composites with the necessary properties [73]. Since carbon fiber fabric may function as a hard substrate in flexible or wearable electronics, incorporating graphene with bismuth-based active materials is a wise strategy to improve electrochemical performance. These methods, however, have drawbacks such poor stacking and low output [74].

Graphene lattices especially (2D) honeycomb lattices. Due to graphene's unique structure, there are significant energy differences between the chemical orbitals of each bond. This is because both energy bands will cross at the Brillouin zone boundary simultaneously, resulting in a cone-shaped energy spectrum. When electrons in the graphene polymer are exposed to an electric field, they must jump through different energy levels [75]. However, copolymer materials, which are semiconductors, have energy differences between bonded molecular orbitals. As electrons move within a polymer, they must pass through an electric field force and energy difference.

In copolymer materials, the presence of this energy difference makes it difficult for electrons to fully bridge the bonds [76]. Because there are fewer obstacles for free electrons or holes to overcome, conductivity is considerably improved by the energy difference between energy bands. Layered crystal networks provide two-dimensional transport channels that are not limited by solid diffusion, allow the use of high-speed intercalation accelerators, and undergo little structural change during the intercalation process. It is occasionally required to shrink the active particles to nanoscale sizes to enable quick charging and high power when solid ion channels have a tendency to be slow [77].

When producing modified graphene materials, scientists took advantage of the hydrothermal and wet-chemical interactions [78] between graphene oxide (GO) and components or tiny molecules that contain nitrogen to synthesize significant volumes of nitrogen-doped graphene. Nanosheets. These materials, however, did not function well in devices, most likely as a result of improper reaction process alterations that exacerbated surface flaws in the completed graphitic structure. The most recent theoretical study indicates that there is no ongoing theoretical research at this time on employing GO functional grouping [79] for stratification. In this paper, we focus on GO functional groups and present a new technique for synthesizing high-quality low-layer graphene from GO in order to investigate and describe its features [80-84].

The research has shown that isolating GO can enhance the potential results and our comprehension of the steps involved in its synthesis [85]. To do this, it is essential to take into account the variety of synthetic processes, complicated chemical interactions, and how various tactics influence GO.

Determining the ideal environment for optimum leaching effectiveness is essential since the generation of GO necessitates a variety of parameters. Particle size, temperature, agitation rate, and leaching acid concentration are a few factors that affect a leaching operation's viability [86].

For example, using the Hummers method (HM), by adding additional KMnO_4 , the researchers were able to produce 80% of the required amount of GO in just 48 hours [87]. However, It was found that natural Gr could be converted into GO via leaching and HM. But their strategy requires extensive testing. Research has been conducted to shorten the oxidation time required for GO synthesis [88]. Aims are being made to create GO from a variety of industrial wastes, including precursors of the graphitic phase, because GO is crucial to the field of materials research. Carbonaceous residues from high-temperature pyrolysis operations are commonly found in various industrial wastes. To remove r/GO and metallic components from industrial or battery waste, many physical and chemical processes must be performed at low temperatures and with little energy. Liquid extraction is used to remove carbonaceous residues from waste, and then different chemical oxidation processes are used to create GO [88]. The main topics of this review are GO from G characterization techniques and graphene synthesis procedures. The experimental environment changed as a result of the synthesis operations. Characterization and its sample applications enable scaling up GO analysis [89,90].

These benefits make graphene a popular material for usage in industrial and commercial settings. In addition to energy conversion, graphene has been found to be beneficial in a range of mechanical, catalytic and electrical devices.

Published research also claims that particles the size of hydrogen ions (H^+) can flow through graphene sheets [91]. Electrostatic interactions between positively and negatively charged ions help disperse protons into the film. Hydrogen produced by water splitting reactions can be directly stored using graphene-based materials. Despite all these advantages, $\text{g-C}_3\text{N}_4$ also has disadvantages that prevent it from being used as a reliable photocatalyst. These disadvantages include their small surface area, delayed surface reaction kinetics, slow charge transfer rates, and lack of active sites. At the Dirac point, the two strips converge. Graphene and $\text{g-C}_3\text{N}_4$'s photocatalytic activity [92] may be altered in a number of ways, including copolymerization, interface engineering, the addition of different functional groups, doping, and hybridization.

High photocatalytic activity was obtained by combining graphene and $\text{g-C}_3\text{N}_4$ with a number of semiconductor materials. In addition to noble metals, supporting transition metal chalcogenides, metal oxides and metal sulfides can improve photocatalytic efficiency [93]. Various techniques were used to increase the surface area of $\text{g-C}_3\text{N}_4$, including chemical oxidation, thermal oxidation, ultrasonic exfoliation, and chemical exfoliation. The efficacy of graphene and $\text{g-C}_3\text{N}_4$ composites as water-splitting photocatalysts is explored in the current work. Non-metallic planar structure graphene

photocatalysts are cheap and have strong photocatalytic activity because they can absorb H_2 well on their surface [94].

Previous research has found that graphene produced using different techniques has different structural characteristics, which affects its chemical properties and limits its uses. Traditional methods of making graphene produce a highly hydrophobic material [95]. Since redeposition is rapid, graphene's surface area and surface activity are diminished, which dramatically lowers other electrochemical activities.

Doped graphene is created, according to early studies, by adding chemicals like N, B, P, and O into the graphene lattice. These heteroatoms can produce electrochemically active compounds on graphene [96]. Previous research has shown that graphene produced using different technologies has different structural characteristics, which can lead to different chemical properties and limit its use. Traditional methods of making graphene produce a highly hydrophobic material. Since graphene is prone to redeposition, its surface area and surface activity are reduced, significantly reducing other electrochemical activities.

Doped graphene is created by introducing molecules like N, B, P, and O to the graphene lattice, according to prior studies [97]. These heteroatoms can produce electrochemically active compounds on the graphene surface. This occurrence takes place when the battery charges. When graphene is employed in supercapacitors, reversible Faradaic redox reaction displays increased specific capacitance and excellent cycle time. The graphene lattice is specifically a (2D) honeycomb lattice. Two cosine-shaped energy bands are formed due to quantum mechanical transitions involving two carbon sub-lattices [98].

Due to graphene's unique structure, there are significant energy differences between the molecular orbitals of individual bonds. This is because the edge of the Brillouin zone is where the two energy bands converge, forming a cone-shaped energy spectrum [99]. The graphene polymer has energy levels that must be traversed by electrons traveling through an electric field. In copolymer materials, there is still an energy difference in the bound molecular orbitals, making them semiconducting. As electrons move within the polymer, they move across this energy gap under the influence of electric field forces. Because of this energy difference, pi electrons cannot freely pass through the bonds in the copolymer material.

The long-term objective is to modify the energy levels of the present electron energy bands by controlling the addition and absorption of electrons. In order to do this, electrons will either be withdrawn from orbitals that are near to one another or introduced to vacant orbitals in the graphene structure. Electrical conductivity considerably increases due to the energy disparity between the bands, which removes barriers to the free passage of electrons or holes [100]. Due to the crystal network's layered nature, high-speed intercalation capacitors may be employed, and solid diffusion is not constrained. To make up for the comparatively sluggish passage of ions through materials, which is required for quick charging and high power, active particles frequently contract

to the nanoscale. Graphene surface formed during charging and discharging [101]. When graphene is used in supercapacitors, the reversible Faradaic redox process has better cycle times and higher specific capacitance.

The unique GO-cellulose nanocomposite created in this work represents a contribution of the research as an effective biosorbent for color removal from wastewater under ideal conditions. Because it is so easy to make, this new biocomposite has significant utility. It holds great promise for large-scale industrial applications due to its process flow, biodegradability [102] possible biodegradability, mild toxicity, biocompatibility, affinity for methylene blue dye, and low cost.

Natural polymers are used to modify the structure and functional groups of GO in order to boost its adsorption capacity since GO lacks the long-chain branching, stable structure, and abundance of negatively charged functional groups that these natural polymers have. According to studies, the biopolymer cellulose increases the structural and functional properties of graphene oxide, making it suitable for a wide range of applications, GO-cellulose composites have recently been used in many processes such as membrane filtration, enzymatic degradation, and adsorption [103].

1.1. History and preparation of GO

In order to create graphite oxide, Benjamin C. Brodie of Oxford University's chemistry department first subjected it to a potassium chlorate and fuming nitric acid solution. A "paper-like foil" with a thickness of 0.05 millimeters, he claims, was created [104]. The Hummers procedure, which uses potassium permanganate (KMnO_4), sodium nitrate (NaNO_3), and sulfate (H_2SO_4), was created in 1957 by

Hummers and Offeman. Even though it is still commonly used today, significant changes are frequently made. Using very pure reactants and solvents, the biggest monolayer GO with a highly intact carbon framework and a low residual impurity concentration may be created. According to the level of oxidation and production technique, graphite oxide has a variety of characteristics. For instance, at the same heating rate, graphite oxide created using the Brodie process frequently has an explosive peeling temperature point that is greater than graphite oxide prepared using the Hummers method; [104] the difference can be up to 100 degrees. The Tang-Lau technique also has the essential benefit of allowing for different thicknesses by altering the growth circumstances (from single to several layers).

1.2. Structure of GO

Several methods have been documented for synthesizing GO using different oxidizing agents, including the Brodie, Staudenmaier, Hofmann, Hummers, and Tour methods. Additionally, at least six differing models have been suggested to explain the structural characteristics of GO, such as the Hofmann–Holst, Ruess, Scholz–Boehm, Nakajima–Matsuo, Lerf–Klinowski, and Zsabó–Dékány models Figure 2 [15]. The structure of GO has been thoroughly investigated, yet there is not a single structural explanation that makes the most sense. The primary causes of this are the complexity of chemicals, the uniqueness of each sample, and its unique stoichiometry [104]. In a nutshell, GO is a single-layer graphite sheet with oxygen groups around its edges and basal plane. The hydroxyl, carboxyl, and epoxy groups are some of these groups. As a result, carbon atoms that are sp^2 and sp^3 hybridized [105].

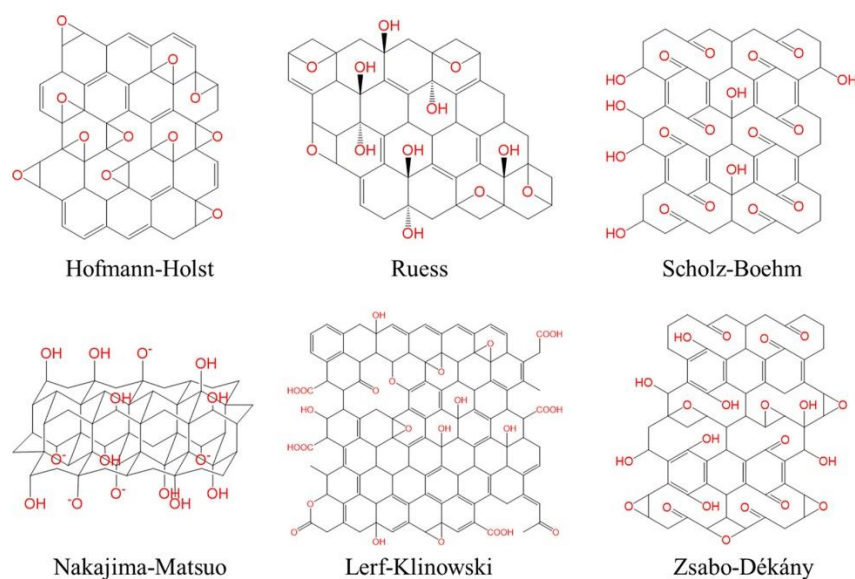


Fig. 2. Six proposed GO models, Reprinted with permission from ref. [15], Tao, W., Lan, Y., Zhang, J., Zhu, L., Liu, Q., Yang, Y., Yang, S., Tian, G. and Zhang, S., 2023. Revealing the Chemical Nature of Functional Groups on Graphene Oxide by Integrating Potentiometric Titration and Ab Initio Calculations. *ACS Omega*, 8(27), pp.24332-24340. Copyright © American Chemical Society.

Many models based on different studies and theoretical simulations have been developed for GO. In 1939, Hofmann and Rudolf proposed the first model when a graphite monolayer is covered by many dispersed epoxy groups [106]. In 1946, Ruess added hydroxyl groups and replaced the sp^2 hybridized carbon in the model with sp^3 hybridized carbon. Scholz and Boem [107] described a less well-organized structure in 1969, i.e., corrugated carbon layers without ether oxygen and occasional C-C single bonds and C=C double bonds. Matsuo and Nakajima [108] later published a comparable model for graphite intercalated compounds in 1994. The LK model was developed by Lerf and Klinowski in 1998 with two distinct sorts of regions: one with a six-membered aliphatic ring and the other with an unoxidized benzene aromatic ring.

The size of these two zones depends on how much the material has oxidized. The top and bottom single layers of carbon are joined by the oxygen functional groups to form two layers of oxygen atoms with different densities. These layers mostly consist of hydroxyl and epoxy groups that are connected by tight bonds. Each oxygen functional group, aromatic body, and oxidized ring are dispersed at random on the carbon monolayer. The acidic character of GO is caused by the hydroxyl and carboxyl groups connected to the lattice's edges. One of the most well-liked and well-known models for the breakdown of oxidized GO is the LK model. Researchers have been studying GO and its derivatives all around the world since the discovery of graphene [108]. Szábó and Dékány offered a carboxylic acid-free model with two primary parts in 2006 after investigating previous models through numerous tests. These sections are trans conjugation with 1,3-epoxides and tertiary alcohols [109]. Different carbon structures generated with accompanying water have

been described using dynamic structural models (DSM), as opposed to static LK models. The observation of C=O bonds at the edge of a carbon monolayer confirmed various aspects of earlier models (especially the LK model) [110]. The chemical composition, and the manufacturing procedure of GO and r/GO has been presented in Figure 3.

Other errors in real GO include adatoms, vacancies, and adsorbed contaminants. Topological defects include pentagons, hexagons, octagons, etc. When concentrated acids are used for oxidation, low molecular weight particles or "oxidized debris" are produced [111-112]. Through interactions including stacking, hydrogen bonding, and van der Waals, highly oxidized PAH fragments (referred to as oxidized fragments) are held to weakly oxidized GO platelets [113]. The number of oxidized debris depends significantly on how long concentrated acid comes in contact with graphite [114-115]. To get rid of these contaminants, some basic cleaning is needed. When compared to chemically reduced GO, pure GO without oxidized debris underwent an equivalent quantity of oxidation [116].

1.3. Properties of GO

Graphene oxide is often called an electrical insulator, just like graphite oxide, due to its broken sp^2 bond network [117]. But that's not entirely accurate. The degree of oxidation of the molecule and the manufacturing process both affect graphene oxide's ability to conduct electrons. Although graphene oxide can transmit electricity, even though its quantities are greatly reduced, it is still not as good at electron mobility as a single layer of high-quality graphene. This is because even after severe oxidation, graphene oxide is still a poor conductor of electricity.

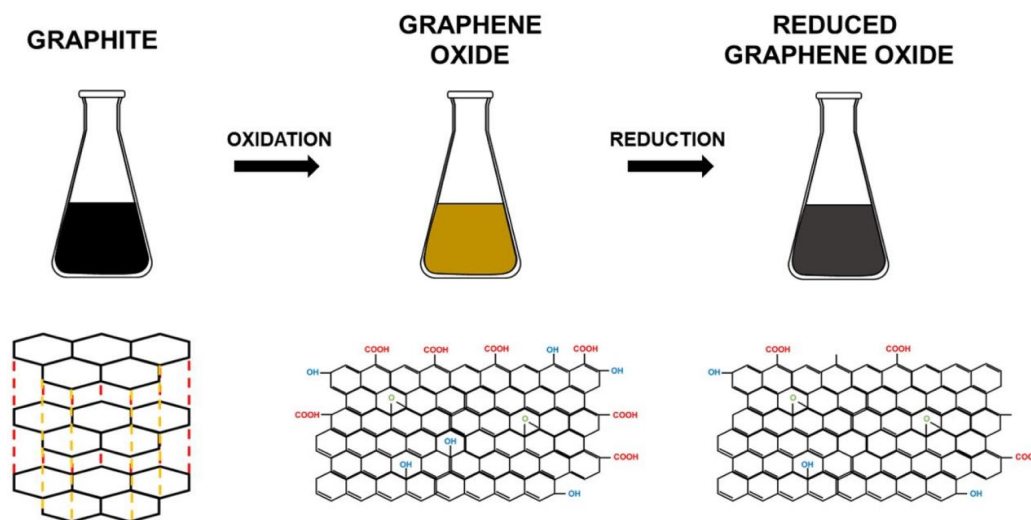


Fig. 3. The chemical composition and manufacturing procedure of GO and rGO. Reprinted with permission from ref. [81], Ghulam, A. N., dos Santos, O. A., Hazeem, L., Pizzorno Backx, B., Bououdina, M., and Bellucci, S., 2022. Graphene oxide (GO) materials-Applications and toxicity on living organisms and environment. *Journal of Functional Biomaterials*, 13, p. 77. Copyright © MDPI.

Table 1. Graphene, GO, and rGO general properties. Reprinted with permission from ref. [64] Taha, M. H. F., Ashraf, H., and Caesarendra, W., 2020. A brief description of cyclic voltammetry transducer-based non-enzymatic glucose biosensor using synthesized graphene electrodes. Applied System Innovation, 3(3), p. 32. Copyright © MDPI.

Properties	Graphene	GO	rGO
Electron Mobility at RT	~200-2500 K ($cm^2V^{-1}S^{-1}$)	0.1-10 ($cm^2V^{-1}S^{-1}$)	2-200 ($cm^2V^{-1}S^{-1}$)
Surface Area	2630 m^2g^{-1}	736.6 m^2g^{-1}	466-758 m^2g^{-1}
Thermal Conductivity	~5000W $m^{-1}K^{-1}$	0.5-18W $m^{-1}K^{-1}$	1390-2275 W $m^{-1}K^{-1}$
C-C Bond Length	0.142 nm	N/A	N/A
Specific Capacitance	550F g^{-1}	215-255F g^{-1}	210-425F g^{-1}
Electrical Conductivity	~ $6 \times 10^8 Sm^{-1}$	5.7 \times $10^{-6} Sm^{-1}$	$10^2 - 10^5 Sm^{-1}$
Sheet Resistance	200 Ωsq^{-1}	~ $10^{10} -$ $10^{12} \Omega sq^{-1}$	~ $10^2 - 10^6 \Omega sq^{-1}$

This is due to the oxidation of the solution affecting the conductivity. However, there are some solutions to this problem, one of which is a process called functionalization [118], which requires chemically changing the properties of substances to make them more useful for specific applications. The main features are categorized as (a) Hydrocystis, (2) Spreadability and (3) Toxicology.

The electrical characteristics of functionalized graphene oxide can be dramatically altered. The resulting chemically modified graphene (CMG) may have many more uses, with almost unlimited uses. Depending on the intended use, graphene oxide can be functionalized through a variety of methods.

The production of well-dispersed metal nanoparticles on both organic and inorganic substrates is demonstrated in this work using the X-ray synthesis of gold (Au), palladium (Pd), and ruthenium (Ru). We synthesized ruthenium (Ru) and palladium (Pd)-based 2D and 3D GO nanostructures. The tiniest particles observed, 3D GO structures, supported on Ru were further researched utilizing an olefination technique as a proof of concept. The fine chemicals sector depends on these methods. The two-dimensional nanomaterial graphene oxide has a high capacity for adsorption, great mechanical strength, and strong thermal conductivity. They are being researched in the fields of sensing, medication delivery, energy storage, fuel cells, and catalysis for these reasons. GO can efficiently catalyze the selective oxidation of CO and alcohols as well as hydrogenation, cross-coupling reactions, fuel synthesis, and impurity eradication. The hunt for more resilient carbon catalysts has been successfully supported by the 3D network of GO, also known as a carbon monolith [114]. Today, many chemical processes utilize metals placed on 2D and 3D GO as catalysts. For instance, Ru is a stable metal at low temperatures, but it changes into various ruthenium (RuOx)-based oxides and hydrated ruthenium oxide (RuO₂·xH₂O) at high temperatures. Due to the deposition of Ru and Ru oxides, mesoporous carbon, carbon nanotubes, and GO all show enhanced conductivity and catalytic activity on these carbon substrates [118]. The

properties of GO has been presented in Table 1.

2. SYNTHESIS OF GRAPHENE OXIDE (GO)

GO is made up of sheets of graphene embellished with hydroxyl and epoxy groups [119]. Due to the pyrolysis of oxygen-containing functional groups to produce gas and the evaporation of interlayer water, rapid heating of GO can cause expansion and delamination. The method of synthesis and the level of oxidation have an impact on the make-up and characteristics of GO. The most used methodology for chemical synthesis is the Hummer method. During this production process, the oxidizing chemical utilized to exfoliate the graphite flakes might change [120]. British scientist B.C. Brodie made the discovery of granulite in 1859 as a consequence of a reaction that took place when KClO₃ was added to a slurry of graphite in fuming HNO₃. A by-product termed "graphic acid" is created during the procedure. Forty years later, L. Staudenmaier added equal amounts of concentrated sulfuric acid and chlorate to the mixture in order to boost the acidity, improving Brodie's KClO₃-fuming HNO₃ technique. These adjustments enhance the craft's usefulness and clarity. Before Hummers and Offeman [26] developed a novel oxidation method utilizing concentrated H₂SO₄, KMnO₄, NaNO₃, and graphite, another 60 years had elapsed. The "Hummer Method" is still commonly used in modern research and testing [121]. In recent years, Hummers' method has been improved in many ways, one of which is the addition of an acidic solution of H₂SO₄ and H₃PO₄ to increase the oxidation efficiency. These r/GO preparation recipes are described in more detail in the next sections. Some of the techniques of converting graphene into GO has been noted in Table 2.

2.1. Hummer's synthesis technique

Using Hummers' technique, graphite is subjected to oxidation to create graphene oxide, as shown in Figure 4 [121].

Table 2. Techniques for converting graphite into graphite oxide. Reprinted with permission from Ref. [105] Compton, O. C., and Nguyen, S. T., 2010. Graphene oxide, highly reduced graphene oxide, and graphene: Versatile building blocks for carbon-based materials. *Small*, 6(6), pp. 711-723. Copyright © Wiley.

Year	Brodie	Staudenmaier	Hummers	Modified Hummers	
	1859	1898	1958	1999	2004
Oxidants	KClO ₃ , HNO ₃	KClO ₃ (or NaClO ₃), HNO ₃ , H ₂ SO ₄	NaNO ₃ , KMnO ₄ , H ₂ SO ₄	Pre-ox: K ₂ S ₂ O ₈ , P ₂ O ₅ , H ₂ SO ₄ Ox: KMnO ₄ , H ₂ SO ₄	NaNO ₃ , KMnO ₄ , H ₂ SO ₄
C:O ratio	2.16 2.28	NA 1.85	2.25	1.3	1.8
Reaction Time	3-4 days 10 h	1-2 days 10 days	≈2 h 9-10 h	6 h pre-ox + 2h ox	≈5days
Intersheet Spacing (Å)	5.95	6.23	6.67	6.9	8.3

The procedure involves several steps. Initially, a 1000 mL volumetric flask is placed in an ice bath (0–5 °C) containing 50 mL of 98% H₂SO₄, 2 g of flaked graphite, and 2 g of NaNO₃, while continually stirring the mixture. After two hours of stirring at this temperature, potassium permanganate (6 g) is progressively added to the suspension. The reaction temperature is carefully kept below 15 °C by controlling the addition rate. The mixture is then removed from the ice bath and stirred regularly for two days at 35 °C until it turns pasty brown. Next, 100 mL of water is gradually stirred in, causing

effervescence that raises the reaction temperature swiftly to 98 °C and changes the hue to brown. An additional 200 mL of water is then added to the still stirring solution. To color the solution yellow and stop the reaction, 10 mL of H₂O₂ is added. The mixture is repeatedly washed with 10% HCl and deionized (DI) water to remove impurities, following multiple centrifugations. Finally, powdered graphene oxide (GO) is produced after vacuum drying at room temperature and filtration.

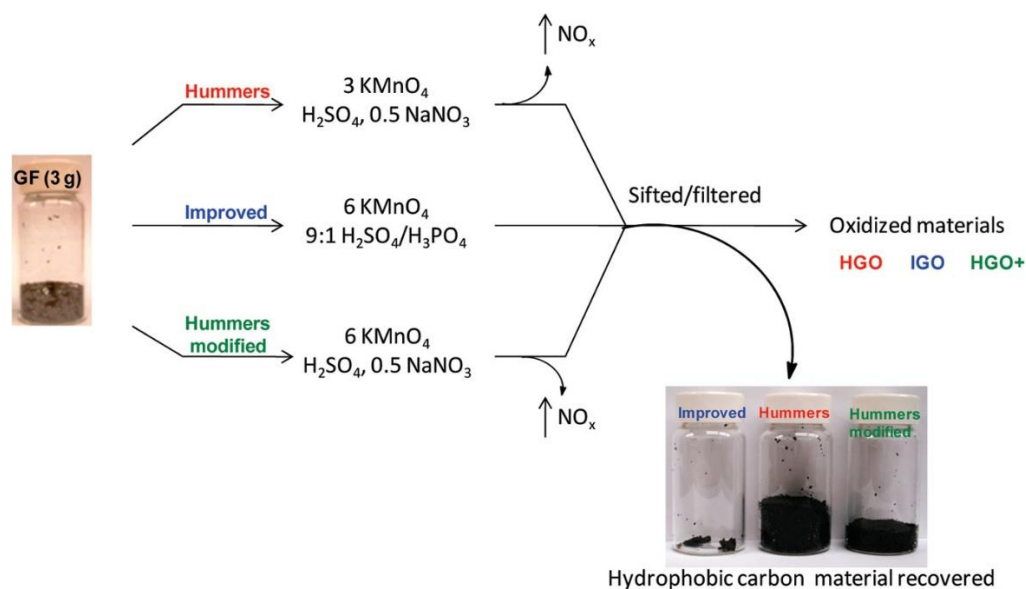


Fig. 4. The Hummer's method for the synthesis of GO. Reprinted with permission from ref. [121], Marcano, D.C., Kosynkin, D.V., Berlin, J.M., Sinitkii, A., Sun, Z., Slesarev, A., Alemany, L.B., Lu, W. and Tour, J.M., 2010. Improved synthesis of graphene oxide. *ACS Nano*, 4(8), pp.4806-4814. Copyright © American Chemical Society.

2.2. Modified Hummer's Method

In the improved synthesis technique, the fluid is heated, causing the graphite flakes to oxidize and flake off. The specific steps of the synthesis process are illustrated in Figure 4. Initially, 2 g of graphite flakes, 2 g of NaNO_3 , and 90 mL of 98% H_2SO_4 are added to a 1000 mL volumetric flask and rotated in an ice bath at 0 to 5 degrees Celsius. After stirring for four hours at this temperature, 12 g of potassium permanganate is gently added to the solution. The addition rate is carefully controlled to maintain the reaction temperature below 15°C . Next, 184 mL of water is gradually added to the mixture, which is then stirred for two hours. The mixture is then agitated at 35°C for an additional two hours after the ice bath is removed. The mixture is kept at 98°C for 10 to 15 minutes in a reflux system. After 10 minutes, the temperature is increased to 30°C to form a brown solution, followed by another temperature change to 25°C , which is held for two hours. Subsequently, 40 mL of H_2O_2 is added to the solution, causing a noticeable yellow change. The resulting solution is divided into two separate beakers, each with 200 mL of water, and stirred for one hour. After standing for 3 to 4 hours, the particles settle to the bottom, and the remaining water is filtered off the top. The resulting mixture is repeatedly centrifuged with 10% HCl and deionized (DI) water to create a pH-neutral gel-like material. Finally, to create GO powder, the gel-like material is centrifuged and vacuum-dried at 60°C for more than 6 hours [4]

2.3. Brodie's method

Nitric acid and potassium chlorate were smoked at 60°C for four days by Brodie, who is credited with producing the first sample of GrO known to science in 1859. Graphic acid was the name he gave to the last chemical, which turned out to be

GO. Due to the use of KClO_3 and the slow recovery, there is an explosion hazard in this method. However, this method leads to oxidation-induced delamination of r/GO sheets into G sheets. Other employees use the procedure to speed up the process and reduce the chance of an explosion.

2.4. Staudenmaier's method

Staudenmaier continued Brody's research, working to improve the safety and yield of surgery. He replaced HNO_3 with a combination of H_2SO_4 and HNO_3 to oxidize r/GO. The Staudenmaier-Hoffman-Hamdi method (SHHM) involves heating potassium chlorate to a high temperature of around 90°C for four days [122] while treating a sand-filled slurry with a solution of sulfuric and nitric acid. This approach increases process efficiency by allowing the oxidation process to occur in only one stage. However, the synthesis process takes four days and is done at high temperatures due to the presence of potassium chlorate, so there is a risk of explosion [123]. Many researchers, including Berthelot, Luzi, Charpy, Weinschenk, Kohlschutter, and Haenni, tried to control the explosive character of the process, but they all failed to produce any useful results by changing the synthesis method [122, 123].

2.5. Tour's approach

Dimiev and Tour enhanced HM by oxidizing rGO without using NaNO_3 using twice the amount of KMnO_4 and an excess of H_2SO_4 and H_3PO_3 (volume ratio of approximately 9:1). This change, now referred to as Tour's modification, reduced the generation of harmful NO_2 and N_2O_4 while doubling the product yield by adding a second reagent, H_3PO_3 [21].

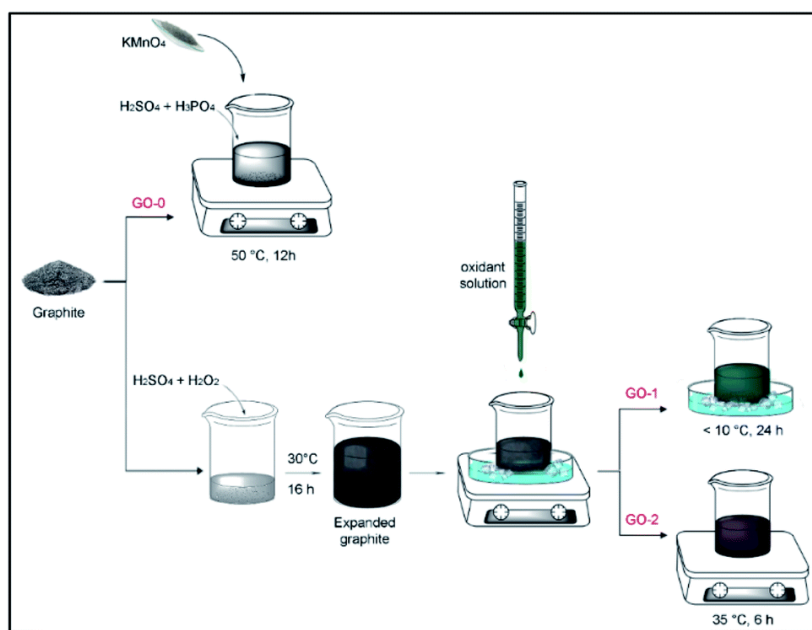


Fig. 5. Schematic illustration of the synthesis of GO. Reprinted with permission from ref. [21], Benzait, Z., Chen, P. and Trabzon, L., 2021. Enhanced synthesis method of graphene oxide. *Nanoscale Advances*, 3(1), pp.223-230. Copyright © RSC.

2.6. Electrochemical Exfoliation

Electrochemical exfoliation has recently evolved into a simple but very successful approach for the industrial synthesis of graphene, shown in Figure 5 [123]. The approach employs a variety of graphite materials as electrodes in aqueous or non-aqueous electrolytes to generate electrode swelling. These materials include graphite powder, graphite plate, graphite rod and graphite foil. Depending on the power input, the electrodes [124] may be cathodic (negative) or anodic (positive) electrodes [125]. Mr. Wang and his collaborators. In their work, they used an electrolyte consisting of PSS (sodium 4-styrenesulfonate) dissolved in deionized water and electrodes made of pure graphite. In an electrochemical cell filled with electrolyte, they placed graphite rods. Use 5 V continuous current. After a few minutes of electrolysis, black matter begins to accumulate on the anode. The stripping process takes 4 hours to extract material from the cells. The material was centrifuged at 1000 rpm and gradually decanted, yielding a stable dispersion [126]. The dispersion was dried under vacuum and weighed after being cleaned with deionized water and ethanol to produce dry graphene in powder form and confirmed from the XRD analysis.

2.7. Brodie's Oxidation Method

By heavily fuming HNO_3 , Brodie pioneered the use of KClO_3 to create a new chemical that combines carbon, oxygen, and hydrogen. This pioneering work improves the quality of flake

graphite. The batch is then rinsed to remove any salt that has formed [127]. During this process, it is dehydrated at 100°C and then put back into the oxidizing atmosphere. After three iterations, a material with a "buff" was formed that was not altered by the additional oxidation treatment. Based on the elemental analysis of his products, he provided the final molecular composition of graphite oxide as $\text{C}_{11}\text{H}_4\text{O}_5$. Furthermore, he found that the chemical precipitated in acidic environments but diffused in pure or alkaline water. Therefore, he named the newly synthesized substance "graphic acid". When heated to 220°C , the material's C:H:O composition changes to 80.13:0.58:19.29 as a result of the reduction of carbonic acid and carbon monoxide. This method can oxidize graphite, but its application is limited by slow response times and potential limitations. Hazardous gas emissions [128].

2.8. Hofmann Method

Since fuming nitric acid is highly toxic and lethal, concentrated sulfuric acid and concentrated nitric acid are used to produce rGO [43, 129]. Due to strong oxidizing power of KClO_3 , acidic solutions often use KClO_3 as an in-situ dioxygen source since it is the primary oxidant. So far, many research teams have successfully made rGO using this technology [130, 131]. The Process diagram by Brodie, Staudenmaier, Hofmann, and HM has been shown in Figure 6 and 7.

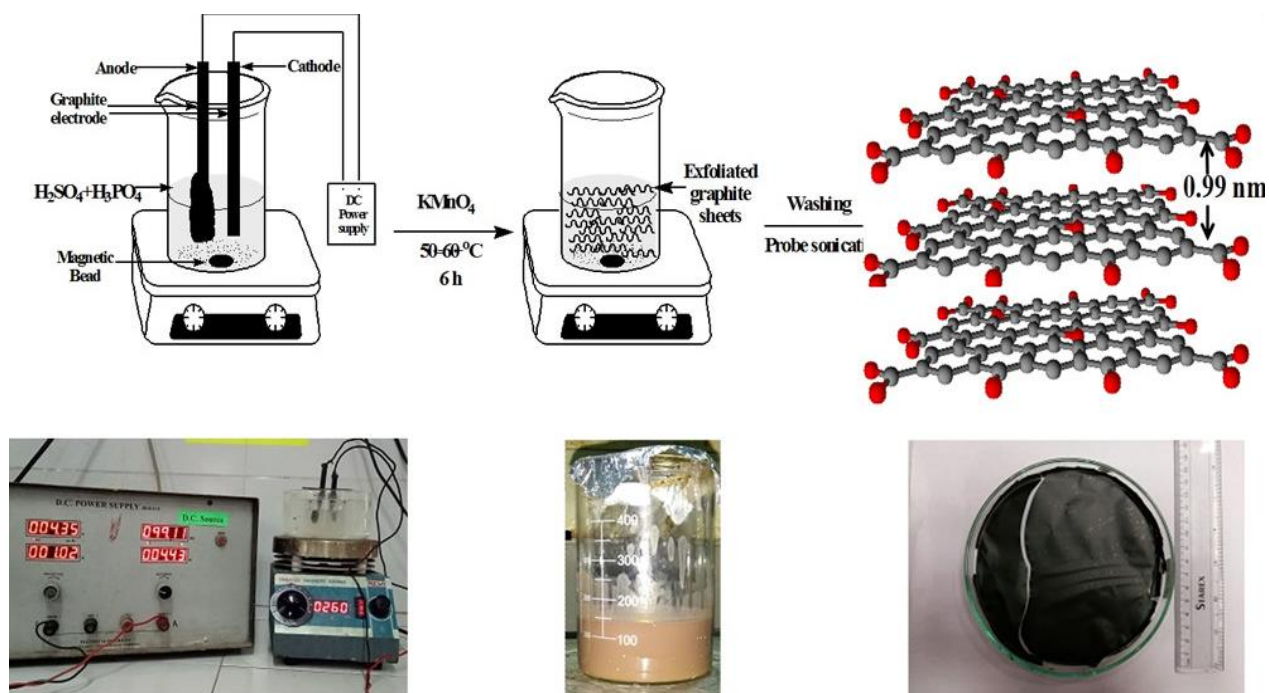


Fig. 6. Simple electrochemical method and oxidation process used to produce large graphene oxide sheets. Reprinted with permission from Ref. [129], Kumar, P., Huo, P., Zhang, R., and Liu, B., 2019. Antibacterial properties of graphene-based nanomaterials. *Nanomaterials*, 9, p. 737. Copyright © MDPI.

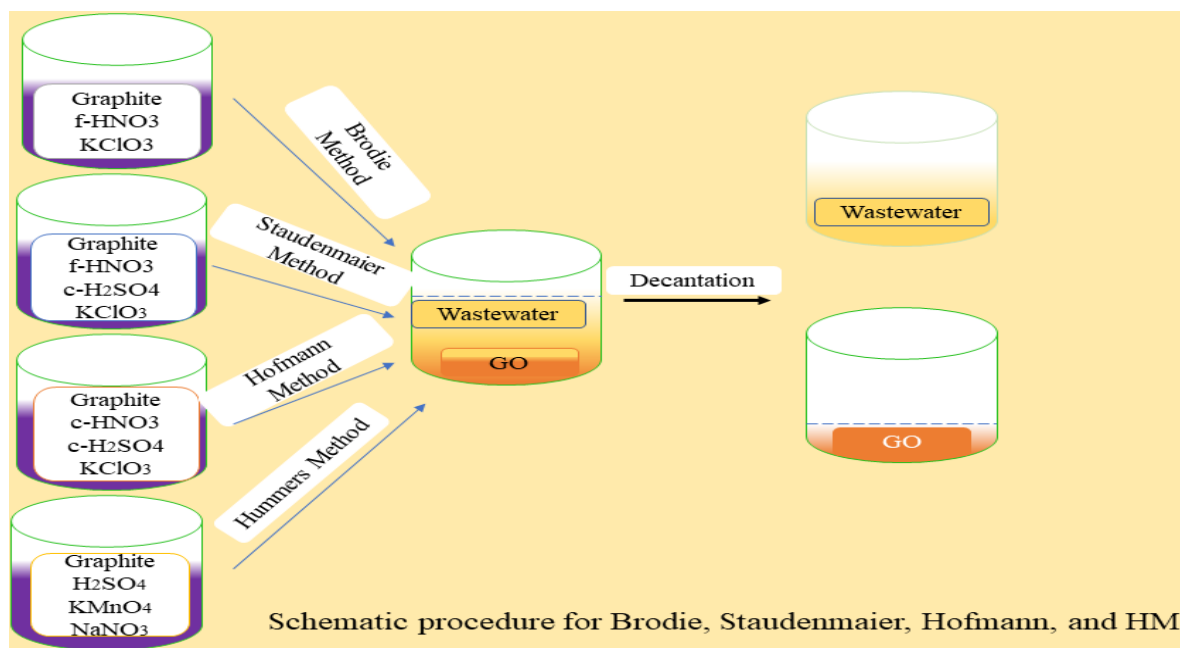


Fig. 7. Process diagram by Brodie, Staudenmaier, Hofmann, and HM. Reprinted with permission from Ref. [136], Le Ba, T., Mahian, O., Wongwises, S., and Szilágyi, I. M., 2020. Review on the recent progress in the preparation and stability of graphene-based nanofluids. *Journal of Thermal Analysis and Calorimetry*, 142, pp. 1145-1172. Copyright © Springer.

3. APPLICATIONS OF GRAPHENE OXIDE (GO)

3.1. Biomedical Applications

The numerous qualities of graphene make it appealing for future biological applications. Due to its substantial surface area, outstanding chemical purity, and simplicity of functionalization, it has the potential to transport drugs. It is the perfect tool for imaging biomolecules in transmission electron microscopy because of its incredible thickness, conductivity, and strength. Graphene that has undergone chemical modification has been used to create testing instruments that are quick and incredibly sensitive [132].

When thinking about possible uses for graphene nanoparticles in biomedicine, biosafety and biocompatibility are crucial factors to take into account. Due to direct interaction with cell membranes or vital internal components, pure graphene nanoparticles demonstrate size-, dose-, and time-dependent toxicity to several cell types. A workable solution to these problems is to modify the physicochemical characteristics of graphene nanoparticles by functionalizing them with biocompatible molecules [133]. With this method, both in vitro and in vivo biocompatibility is increased while cytotoxicity is decreased. Graphene that has been amine-modified (G-NH₂). It was recognized as the most biocompatible and safest graphene derivative in 2012, with the ability to drastically reduce cell toxicity. The impact of GO on the toxicity and aggregation of thrombi. Perini and colleagues have discovered that NH₂-GQDs had the greatest biocompatibility in terms of cytotoxicity and effects on cell membrane permeability when compared to COOH-GQDs

and GQDs. When compared to GO, amino-functionalized GO significantly boosted the number of signaling pathways it activated [134]. Aminated GO, which ranges in size from 280 nm to 6.4 μm, is more hazardous to human liver cancer cells than GO with larger diameters. The range of aminated GO's particle size distribution is 102 nm to 1.944 μm [135]. These biopolymers were chosen because of their exceptional qualities, such as nontoxicity, biocompatibility, and biodegradability, for the creation of graphene-based drug delivery systems. Biopolymers can improve the biocompatibility, cytotoxicity, and stability of graphene nanoparticles [136].

The development of prosthetic nerves, repair of damaged neural tissue, and regeneration of damaged nerves, especially in the spinal cord, can all be enabled by graphene-based nano-medicines for disease diagnosis and treatment. It could be used in bioelectronics limbs as well as retinal or eye materials for blind people [137]. Pure and functionalized graphene layers and its derivatives have lately been advocated as leading contenders for a number of biomedical engineering and biotechnology applications due to their flexible physical, mechanical, and biocompatibility qualities. For instance, because of their superior tensile strength and Young's modulus, polylactic acid derivatives incorporating graphene are used to make surgical implants [138].

3.2. Drug carrying and target delivery

One of the effective therapeutic alternatives being investigated by several researchers for drug delivery based on

graphene materials, using graphene oxide conjugated with hyaluronic acid. In line with this hypothesis, HeLa/L929 cells were effectively treated with drugs. In this experiment, the drug used was doxorubicin (DOX). The drug's formulation uses hyaluronic acid and graphene oxide to help distribute the drug effectively to its target [139]. The findings demonstrate that graphene-HA is able to deliver the drug doxorubicin to tumors while preventing tumor development.

Graphene oxide as a drug transporter targeting HepG2 cells/PEF/HEK293A cells. The Hummers process is employed to manufacture graphene oxide, and SEM, TEM, and XRD are utilized to validate the method's efficiency. Graphene oxide can be used as a transport vehicle to deliver drugs to cancerous tissue. There's no doubt that drug therapy is a successful technique in treating cancer, [140] but some studies suggest it's also successful in treating Parkinson's disease. Puerarin then crosses the blood-brain barrier in the brain via lactoferrin, a specifically targeted ligand. Therefore, the proposed treatment strategy for Parkinson's disease by lactoferrin, graphene oxide, and perillin is successful [141].

GO, an oxidized version of graphene, is the finest material for labeling and extracting different drugs and biomolecules. The enormous surface area and exposed functional groups (epoxy, hydroxyl, carboxyl, carbonyl, etc.) of GO allow for the attachment of both organic and inorganic molecules. GO is frequently used in the administration of pharmaceuticals and biological macromolecules like proteins, peptides, or nucleic acids [142]. GO nanocarriers are able to transport a variety of medications due to their enormous specific surface area, conjugated structure, and considerable surface activity brought on by several oxygen-containing functional groups. CS, a naturally occurring cationic polyelectrolyte, will become positively charged when interacting with the acidic extracellular environment of solid tumors because negatively charged tumor cell membranes are more easily pulled to positively charged nanocarriers. To enable controlled dispersion and long-term sustained release, anti-tumor drugs and biomacromolecules are effectively immobilized or loaded into nanocomposites based on CS and GO. Nanoparticles and hydrogels are now the two most used CS-GO delivery methods [143].

3.3. Bioimaging and biosensing

Examples of medical devices that use biosensors to determine the presence and concentration of chemical compounds include pregnancy tests and blood glucose monitors. They are very effective due to their typical portability, responsiveness and sensitivity [144]. Gold, silver and more recently graphene are all materials used in biosensors. Graphene-based materials, such graphene oxide (GO), have generated a lot of attention in biosensing applications due to their robust signal output and potential for rapid industrial expansion. Strong mechanical attributes (such hardness and flexibility) and high chemical reactivity are only a couple of the many advantageous characteristics of graphene. The presence of surface ripples (whether naturally occurring or artificially

produced) presents another quality or variable with great potential if used appropriately [145].

Around the world, scientists have used graphene and similar nanomaterials to create biosensors capable of detecting a variety of diseases. For example, an immunobiosensor made of graphene oxide can be detected within detection limits between 103 and 105 pfum11. By synthesizing graphene oxide using a modified Hummer method [146] and depositing it onto an amino-modified glass surface, a more sensitive and focused sensor can be produced. In another study, biocompatible-GO was used to create a dependable glucose biosensor with outstanding sensitivity (8.045 mA.cm² M⁻¹) and a linear range of 28 mM.mm². The biocompatibility of manufactured GO nanosheets with human cells (RPE or retinal pigment epithelial cells) was effectively determined for the first time in this work.

Graphene oxide was added to the biosensor along with glucose oxidase [148]. This improves the biosensor's capacity for data storage and measurement repetition. Construction of biosensors supported by nanomaterials that are better able to recognize harmful germs spread in food has become possible because to the rapid advancement of nanoscience and nanotechnology. Reduced graphene oxide (r/GO) nanocomposites are well suited for electrochemical biosensors because to their high specific surface area, outstanding thermal stability, superior electrical and thermal conductivity, high carrier mobility, and great mechanical properties. These elements have had a substantial impact on the development of r/GO nanocomposites-based electrochemical biosensors over time [149]. The GO-CS and MBP-rPC-based glucose sensing biosensor schematic is shown in Figure 8.

Under UV light (ex = 365 nm), K-doped GO is distinctive in that it displays vivid blue photoluminescence (PL). It is made using an affordable, secure, and environmentally friendly synthetic approach from agricultural waste (acorns). The synthesized K-doped GO is a promising bioimaging and biosensing reagent with significant promise in the analytical and biological domains [150] due to characteristics including excellent PL properties, high biocompatibility, intracellular localization, and low toxicity.

GQDs' optoelectronic characteristics and possible applications have lately drawn a lot of interest. Due to their advantageous qualities in biomedical applications, such as photoluminescence, wide surface area, high water solubility, strong biocompatibility, and low cytotoxicity [151], they have lately been employed in bioimaging and biosensing.

We carefully analyzed and assessed the optical and electrical characteristics of CS- and GO-based nanocomposites. CS-modified GO was created especially for FRET (fluorescence resonance energy transfer) glucose detection [152]. The technology (fi) for biosensor-based glucose detection is displayed. Maltose-binding protein (MBP) is marked by the far-red fluorescence produced by the component of recombinant phycocyanin (rPC). The 0.05 mg/mL glucose detection cutoff was used.

3.4. Bone and tissue engineering

The idea that CS-based composites might hasten patient recovery, repair injured tissue, and save expenditures is backed by a number of research. According to current research, CS-GO nanocomposites could one day be used to repair a variety of tissues, including bone, skin, heart, neurons, and other electrically active [153]. A large portion of this research is devoted to bone tissue engineering, with only a small portion focused on other tissues. CS-GO nanocomposites are mainly used to produce bone tissue due to the molecular similarities between CS and glycosaminoglycans (an important component of the bone extracellular matrix), and the ability of GO to enhance osteogenic development when mixed with CS. The created CS-GO nanocomposites are frequently 3D matrices (such as scaffolds and hydrogels) that encourage the ingrowth of new bone tissue by simulating the microstructure of native bone [154].

Low back pain is significantly influenced by intervertebral disc degeneration (IVD), which is a significant example of a condition that might benefit from minimally invasive cell treatment. The IVD is made up of the annulus fibrosus (AF), a rough fibrous tissue, and the nucleus pulposus [154] (NP), an inner gel core. At NP, cell-driven IVD denaturation starts to take place. NP cells release catabolic and degradative enzymes in response to modifications in the physiochemical environment, which lead to tissue breakdown, loss of disc height, and ultimately pain.

Due to the low mitotic activity of NP cells and the

avascular nature of NP, IVD cannot mend itself. Several organizations are presently looking towards cell-based therapies that need little to no invasiveness in order to enhance the quantity of NP cells and regenerate NPs. For instance, a recent randomized clinical research on humans showed that NP cell transplantation dramatically decreased low back pain and preserved disc height and moisture levels while avoiding IVD degeneration in a canine model.

The use of injectable hydrogels to transport cells to NPs has generated interest [155]. In fact, hydrogels can retain and/or restore the mechanical functioning of nanoparticles in addition to enabling cell dispersion and preservation. Additionally, they enable replication of the mechanical characteristics and highly hydrated condition of NPs. Due to their biocompatibility and lack of immunogenic potential, peptide-based hydrogels are a unique class of hydrogels that have garnered significant interest in the last ten years. To make peptide hydrogels for a range of biological applications, including injectable self-healing cell delivery systems, several peptide patterns have been developed. A hydrogel is made of fibers that entwine or connect to form a three-dimensional network of fibrils that are inflated by water [155]. These hydrogels have demonstrated that they are effective for growing NP cells as well as other cell types. In order to customize the characteristics and usefulness of peptide-based hydrogels, new technology is being used with nanofillers, particularly carbon-based nanofillers. Due to its high water dispersibility, outstanding biocompatibility, [156] and characteristics that encourage cell attachment, graphene oxide (GO) is exceptional.

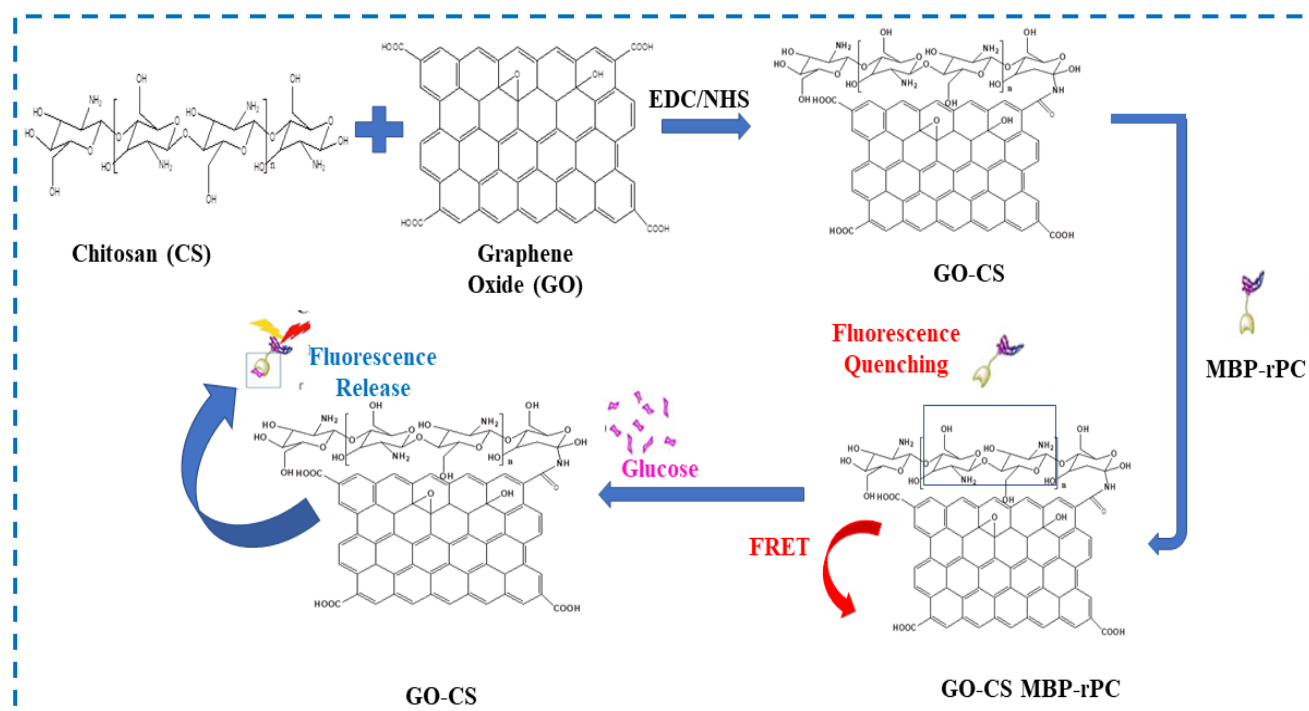


Fig. 8. GO-CS and MBP-rPC-based glucose sensing biosensor schematic. Reprinted with permission from Ref. [139], Li, W., Jiang, T., Pu, Y., Jiao, X., Tan, W., and Qin, S., 2019. Glucose biosensor using fluorescence quenching with chitosan-modified graphene oxide. *Micro & Nano Letters*, 14, pp. 344-348. Copyright © Wiley.

Since there are so many functionalization techniques available, GO is also a potential mediator for the functionalization of peptide hydrogels [157]. The creation of hybrid peptide hydrogels for the transportation of NP cells utilizing GO as a nanofiller was the focus of this investigation. To do this, we first added GO to the hydrogel and examined its physicochemical properties. NP cells were subsequently contained and cultivated for 7 days to demonstrate the potential utility of hybrid hydrogels as scaffolds for their transport [157].

The creation of hybrid peptide hydrogels for the transportation of NP cells utilizing GO as a nanofiller was the focus of this investigation. To do this, we first added GO to the hydrogel and examined its physicochemical properties. NP cells were subsequently contained and cultivated for 7 days to demonstrate the potential utility of hybrid hydrogels as scaffolds for their transport [157].

3.5. Chemotherapy

Although photothermal therapy (PTT) has become a cutting-edge method for treating patients, tumor chemotherapy still has many limitations. In PTT, photothermal agents are used, either directly injected into the diseased area or as a targeting agent [158]. When relaxed, these photothermal agents produce heat that is damaging to cancer cells because they are selectively triggered by near-infrared (NIR) light. Theranostic systems, or nanosystems that manage detection and therapy and are expected to be helpful in eliminating cancer cells, have been the subject of much research. Numerous photothermal agents, such as organic, inorganic, organic-inorganic, and their composite materials, have been researched thus far [159]. Carbon nanomaterials, in particular carbon nanotubes, graphene, and graphene derivatives, are well known for their exceptional intrinsic qualities and distinctive chemical make-up. They can be used in a variety of processes, including photocatalysis, electromagnetic shielding, electrical applications, and fingerprint recognition. The biomedical engineering disciplines of tissue engineering, drug delivery, biosensing, bioimaging, and cancer therapy and diagnostics have benefited greatly from these carbon nanomaterials. For the past 10 years, graphene oxide (GO), also known as oxidized graphite, has been the focus of much study [160]. The fact that graphene oxide dissolves readily in water as compared to graphene has encouraged study into potential biological applications. Chemical alterations are required to make graphene water-soluble and appropriate for extracellular and intracellular settings. Due to the employment of dangerous and deadly chemicals during their production and surface modification, GO sheets are intrinsically damaging. These molecules may still be present in the material in tiny concentrations, [161] making it potentially dangerous both within and outside the body. Only a small amount of natural graphene oxide is luminous. To fully exploit these nanomaterials' potential for usage in diagnostics, theranostics, and biomedical optics, it is essential to connect them with organic dyes/imaging probes to locate them in extracellular and intracellular contexts. These dyes

are limited in usage and unable to fulfill their intended function due to three key issues: photostability, flicker, and environmental sensitivity. We address these challenges using graphene quantum dots (GQDs), a zero-dimensional graphene oxide derivative with excellent physical and chemical properties, including high fluorescence, photostability, and biocompatibility. Given their straightforward production utilizing a variety of carbon-based precursors, GQDs are commonly employed in bioimaging and photothermal reactions [162]. We used polyethyleneimine (PEI) as a linker to functionalize GO sheets by taking use of these characteristics of GQDs. A cationic polymer called PEI has been employed in earlier research for uses including enhancing fluorescence, biosensing, linking two nanostructures or moieties, and gene transfer. PEI's positive charge aids in bringing together the two negatively charged moieties to create a stable hybrid.

The decrease of cells in the presence of MTT has been used to test the cytotoxicity of r/GO against cancer cell lines such as A549 and HepG₂. The cytotoxic mechanism of r/GO against cancer cell lines may involve extracellular matrix or plasma membrane [163]. They often do this via binding to receptors, undergoing endocytosis, or spreading throughout the cell. The key determinants of graphene's toxicity are its physical characteristics, its interactions with cells, and how much of it accumulates in a given organ. When graphene interacts with light, reactive oxygen species may lead to oxidative stress, cellular malfunction, pro-inflammatory responses, and mitochondrial damage. The transport of graphene into the nucleus may lead to DNA strand breakage, the activation of transcription factors, the stimulation of gene expression, cell death, and genotoxicity [164].

In the nonpolar (organic) chemical chloroform, PEG-grafted TOCNCs display homogeneous dispersion at the nanoscale element level. PLA/PEG-TOCNC/rGO nanocomposites films were created by solvent-casting a mixture of PLA/chloroform solution, PEG-grafted TOCNC/chloroform solution, and r/GO/chloroform solution onto a Petri plate. Both tensile strength and thermal stability were increased in films with the greatest weight % modified CNC. Because it still exhibits some antioxidant activity and water vapor barrier capabilities, the film can be employed as a packaging film [165]. Additionally, the movie is practically nontoxic to C₃H₁₀T_{1/2} fibroblasts and cytocompatible. Due to its advantageous characteristics, PLA/PEG-TOCNC/rGO nanocomposite membranes may be employed as packaging materials as well as tissue engineering scaffolds [165]. Copper oxide (CuO) nanoparticles have received a lot of interest due to their unique physical and chemical properties. Only a few of its distinctive qualities include a sizable specific surface area, strong electrochemical activity, an appropriate redox potential, and good solution stability. Similar to the subject of precious metal nanoparticles, it is intriguing. The use of copper oxide nanoparticles in research has significantly risen since nanotechnology was introduced to the biomedical sector [166]. Since CuO nanoparticles have a large specific surface area and may speed up the electron transfer process at low overpotentials, they are frequently

utilized for non-enzymatic sensing of therapeutically relevant analytes. CuO nanoparticles have been shown to have pharmacological effects, particularly when utilized to treat cancer. The effectiveness of copper oxide may be increased in a number of ways, but mixing it with carbonaceous substances like graphene oxide (GO) in the form of nanocomposites has garnered a lot of attention [166]. These materials are anticipated to be a key route for enhancing CuO decomposition activity due to their enormous surface area, high thermal conductivity, rapid charge mobility, and photostability. CuO nanoparticles alone offer less benefits than GO and CuO nanocomposites, which also increase the surface area of CuO and enhance its adsorption capability. In order to hasten the healing of wounds, GO/CuO nanocomposites (NCs) generated by the sol-gel technique are essential. These NCs can encourage accumulative cell proliferation, angiogenic processes, and the quick beginning of inflammatory responses [167].

3.6. Desalination and Water treatment

Desalination is the process of extracting dissolved minerals from salt water, including seawater, brackish water, and treated water [168]. Traditional seawater desalination methods include membrane filtration, ion exchange, precipitation, oxidation, reduction and adsorption. Adsorption is one of the most promising and popular methods when considering the ease of use, cost-effectiveness, and popularity of the process. Adsorption is directly influenced by the characteristics of the utilized adsorbent, such as its specific surface area and surface functional groups (active adsorption sites). In terms of sodium ion adsorption, the modified Hummers strategy performs better than the original Hummers strategy [169]. Additionally, chitosan functionalization improved the modified Hummers method-fabricated GO's capacity to adsorb sodium ions. Carbon nanomaterials including graphene oxide (GO), carbon nanotubes (CNT), and graphene and its derivatives have drawn a lot of attention as prospective adsorption materials because of their distinct physical and chemical characteristics.

Hexagonal planar monolayers of sp² hybridized carbon atoms make up the two-dimensional carbon-based nanomaterial known as graphene [169]. Specific oxidation techniques must be used to introduce hydrophilic functional groups into graphene to improve its ability to adsorb ions. By oxidizing graphite, graphene oxide, called GO, can be produced. Oxidation of graphite causes oxygen molecules to spread over the carbon layer. Graphite oxide becomes graphite oxide (GO) through exfoliation, thereby expanding the interlayer gap [170]. Two examples of the many oxygen-containing functional groups that the Hummers and modified Hummers processes introduced to the GO surface are the carboxylic acid groups at the GO borders and the hydroxyl and epoxy groups on the basal planes. The hydrophilicity and high negative charge density of GO are significantly increased by these oxygen-containing functional groups, two properties that are crucial for desalination applications.

Activated carbon (AC), fly ash, sewage sludge ash, zeolites, biomaterials, recovered alum sludge, manganese oxide, peanut shells, kaolinite, resins, rice husk, clay, sawdust, and carbon nanotubes are some more adsorbents that may be utilized to adsorb various chemicals. However, GO is thought to be the most promising adsorbent because to its unique physical features [171]. GO must possess a large amount of adsorption capacity in order to effectively remove sodium ions from salt water during desalination. The removal of waste GO from water can be made easier by functionalizing GO, according to results in the literature, which also increases its adsorption capability and selectivity. The number of active adsorption sites in GO, for instance, may be greatly increased by the addition of EDTA groups during silanization, producing EDTA-GO with more sites than GO [172].

Due to their high water permeability and stability in water, GO membranes are frequently thought of as possible materials for use in water treatment. The creation of high permeability membranes with high rejection rates has made great strides in recent years [173]. The GO sheets were produced on polydopamine-treated PES scaffolds when tannins and theanine were cross-linked. When GO was added to polyamide at a concentration of 0.2 weight percent, the resulting GO/polyamide nanocomposites increased water flow through alkaline polyamide membranes by a factor of 12 while still maintaining high salt rejection [174].

GO membranes frequently encounter issues as a result of the several procedures used in desalination and other methods of water treatment. In dry and wet settings, the GO interlayer spacing may rise from 7.76 to 17.63, respectively. It is challenging for the GO membrane to reject ordinary salt because the hydrated ions' diameter is lower than this hydration value [175]. GO nanosheets (fi A-D) were physically confined and encased in epoxy resin at various relative humidity levels, we were able to manufacture GO films with lower interlayer distances while retaining interlayer distances of 9.8-6.4. The amount of salt that can pass through the membrane at the chosen interlayer spacing is quite minimal, as can be observed by how quickly it does so. Less permeation results with smaller interlayer spacing because at 6.4, there is no observable ion concentration in the permeate. According to the aforementioned results, GO nanosheet interlayer spacing may be carefully adjusted to retain significant salt rejection [6]. The epoxy appears bright yellow with black streaks due to surface flaws [176]. (D) A scanning electron micrograph taken in the area that was given the C label. Scale bar of 1 mm. The interlayer spacing in (E) (inset), which is sensitive to humidity, was found using X-ray diffraction. Similar examples were given to illustrate the issue with liquid water. The standard deviation determined from at least two measurements of three separate samples is shown by error bars. (F) PCGO membranes with varying interlayer distances and color-coded permeability. Salts included KCl, NaCl, LiCl, CaCl₂, and MgCl₂. Published data were used to calculate hydrated diameter [118]. As a visual clue, the dashed line shows the interlayer spacing, which influences salt permeability. For our five days of data, the

gray region is below detection limits, with arrows pointing to limitations for certain salts. Our detection threshold for Cl is represented by the horizontal blue line. Above the latter limit, cations and anions are present in stoichiometric amounts.

3.7. Cleaning up oil contamination in water

Treating oily industrial wastewater before discharging it into the environment or using it for other purposes is one of the most difficult tasks worldwide today [176]. Many industries such as metallurgy, pharmaceuticals, petrochemicals, and oil refining produce large amounts of wastewater every day, causing harm to the environment. Water pollution is increasingly recognized for its ability to damage ecosystems. It is difficult to distinguish emulsified oil from effluent due to the tiny size of the oil droplets [177]. Gravity sedimentation, flotation, coagulation, adsorption, biological treatment, and advanced oxidation processes are just a few of the methods used to treat oily emulsion wastewater. Membrane filtration technology is more efficient and gives a potential future solution for successful oil-water separation since it is based on a "size sieving" process that allows the entrance of small water molecules through the membrane while rejecting bigger contaminants. In a recent study, gasoline was effectively extracted from an oil/water emulsion that had an excessive amount of salt using graphene oxide. GO has a larger potential for oil/water separation than pristine graphene because its surface contains more oxygen functional groups than pure graphene. Additionally, GO nanosheets self-assemble on the microporous substrate during the vacuum filtering process, [178] forming nanochannels between the sheets that only let water molecules to pass through. When used on polymeric microporous membranes, the hydrophilicity and antibacterial capacity of GO also demonstrate antifouling capabilities. The collection of oil, bacteria, and inorganic particles, which hinder the membrane's capacity to carry water, are the major causes of membrane fouling [179]. It is advised to use a hydrophilic substance to cover the polymer membrane in order to address this issue since it forms a hydrated layer and prevents impurities from sticking to it.

The disadvantage of pure GO membranes is that they enlarge throughout the filtering process, allowing more space between GO sheets and enhancing the ability of petroleum contaminants to flow through the membrane. To increase the durability of graphene oxide nanosheets, it is desirable to chemically modify them prior to placement [180].

Tannic acid cross-linked GO membranes have been created for effective oil-contaminated water filtration. Tannic acid is a naturally occurring chemical that interacts electrostatically with the functional groups of graphene oxide thanks to its many terminal phenolic hydroxyl groups. To hold the coating in place, tannic acid molecules even create non-covalent connections with the PVDF membrane [181]. In comparison to other hydrophilic coatings like polydopamine, which block the membrane's pores and drastically reduce flux, TA coating may dramatically boost

the water flow during filtering. Membranes made from these cross-linked GOs are hence very strong and fouling-resistant. Tannic acid gives the membrane a thick layer of hydroxyl groups on its surface, making it hydrophilic [182]. When there are additional pollutants in the water in addition to oil, this membrane is not suited for wastewater filtering. A tannic acid cross-linked membrane is advantageous in these circumstances because it permits only water molecules to pass through, producing clean water. This improved technology will be more effective overall and lower treatment technology costs by addressing the aforementioned restrictions [183].

3.8. Pollutant degradation by photocatalysis

Infections such as bacteria, viruses, and dyes often contaminate the environment and endanger humans [184]. Organic dyes are hazardous compounds that may be present in various industrial wastewaters. Examples include indigo carmine (IC, anionic), methylene blue (MB, cationic), etc [185]. Due to its capacity to utilize a variety of plentiful natural light spectrum to break down chemical (dyes) and biological (pathogens) contaminants, visible light-active semiconductor photocatalysis research is of exceptional academic and ecological relevance [186]. As light reactions are not always created purely by exposure to light, photocatalysts are molecules that are synthesized with the help of photons and are catalytically active [187]. A semiconductor material begins to photocatalyze when exposed to visible or ultraviolet light, which causes electrons to be lifted from the valence band to the conduction band [187]. On the surfaces of the nanoparticles, these holes and electrons also participate in oxidation and reduction processes [187]. Due to its narrow band gap (2.4 to 2.8 eV), tungsten oxide (WO_3) is an n-type semiconductor with potential photocatalytic effect under visible light irradiation and is one of the most promising semiconductor photocatalysts for destroying organic molecules [188]. It is reasonably priced and has good stability in acidic aqueous solutions [188]. A recent study that showed the metal's safety and outstanding biophotocatalytic capabilities has encouraged more research into tungsten oxide's potential use in nanobiotechnology [189]. The inefficiency of WO_3 as a photocatalyst is due to the fact that the photogenerated electrons on it are unable to decrease the absorbed O_2 , which leads to the recombination of electrons and holes on its surface [190]. It has been demonstrated that the photocatalytic activity of WO_3 is enhanced by the capacity of noble metals like Ag, Pt, and Au to effectively absorb WO_3 electrons and avoid recombination [191].

Since precious metals are expensive and rare, great effort has gone into developing a less expensive substitute. As a result, tungsten oxide's photocatalytic activity rises as well [191]. Amazing features of graphene, including a sizable specific surface area, outstanding electrical conductivity, quick carrier mobility, and high mechanical properties, have sparked a substantial amount of interest in graphene-based

composites among researchers [192]. Additionally, it significantly boosts photocatalytic activity. Defect sites in GO may trap excited carriers and stop recombination. The intriguing property of graphene to transport electrons has inspired study into the development of semiconductor photocatalysts based on graphene and how they might be used to alter the environment pollutants [193]. Medical professionals often use antibiotics to treat microbiological illnesses in both people and animals. They are further added to feed to help agricultural animals flourish [194]. Antibiotics are widely detected in a range of habitats, including terrestrial and aquatic settings, surface groundwater, and direct drinking water, as a result of their ubiquitous usage [195]. There is no denying that antibiotics have numerous medicinal advantages, but excessive usage can threaten the environment by accelerating bacterial resistance and rendering even the most widely used medications ineffective for treating infectious illnesses. Furthermore, even at incredibly low concentrations, there are significant worries regarding their toxicity to creatures that are not the target. Antibiotic parent chemical residues ultimately leave the body through urine and feces and are carried to wastewater treatment facilities because antibiotics are not metabolized in the body [196]. As a result, several techniques have been investigated to remove antibiotics from wastewater, including biological treatment, disinfection, membrane filtration, electrochemical treatment, and activated carbon adsorption. These systems do have several shortcomings, though, including expensive costs, limited removal power, and difficult response environments [197].

Due to photocatalysts' production of reactive groups like OH and O₂, antibiotics can successfully break down or degrade when exposed to sunshine, visible light, or ultraviolet (UV) radiation. Therefore, one of the most effective, environmentally friendly, and well-liked strategies for eliminating antibiotic pollutants from the environment is photocatalytic degradation [198]. The promising photocatalyst graphene has been intensively studied for its ability to remove antibiotics from wastewater [199]. Due to its high intrinsic electron mobility, superior electrical conductivity, optical transparency, and strong chemical stability, graphene would be a wise candidate for photocatalytic applications. Graphene can function as a cocatalyst by extending the range of light that can be absorbed thanks to its polymer adsorption capability, in addition to being a good electron acceptor that avoids the recombination of photogenerated electron-hole pairs [200]. However, when graphene sheets start to aggregate, they easily lose their catalytic activity. Graphene can decompose antibiotics due to its interaction with other semiconductor photocatalysts [201].

3.9. Adsorption of pollutants

Around the world, environmental toxins such heavy metals, organic pollutants, and nutrients are a critical concern since they may adversely affect human health either directly or indirectly [201]. The neurological, skin, respiratory, blood,

and liver systems can all be harmed by heavy metals, which include mercury, cadmium, arsenic, lead, chromium, antimony, thallium, copper, zinc, nickel, selenium, cobalt, and molybdenum. Brings on cancer. The major sources of heavy metals include sewage, industrial waste, and the mining and smelting of metal resources [202]. The ecosystem is harmed by these contaminants because they are hard to degrade and can biomagnify via the food chain. Some heavy metals, such Cr, As, Cd, Hg, and Pb, frequently bind to thiol groups (-SH) in enzymes that control the rate of metabolic processes, impairing their functionality [202].

Numerous organic contaminants are teratogenic, endocrine disruptors, carcinogens, and mutagenic. Organochlorine insecticides, polybrominated diphenyl ethers, polychlorinated dibenzofurans, polychlorinated dibenzo-p-dioxins, and perfluorinated chemicals are examples of persistent organic pollutants. These pollutants are particularly harmful due to their high toxicity, difficulty in degrading, easy absorption by organisms, and wide diffusion [203]. Water contains inorganic substances such as silicon, phosphorus, and nitrogen, also called "nutrients." Season, water depth and biological activity all have significant effects on its concentration [204]. Eutrophication can occur if the nutrient content in a body of water becomes too high. Nitrogen, phosphorus and other nutrients are introduced as the quality of slow-moving water bodies such as rivers, lakes and bays decreases, dissolved oxygen levels decrease, and fish and other species become decimated [205]. Due to its ease of use, low cost, and simplicity, adsorption now has a wide range of potential applications. The three main forms of adsorption, based on adsorption capacity, are physical, chemical, and ion exchange. Contaminants can be absorbed through a variety of methods, such as ionic bonds, covalent bonds, hydrophobic contacts, hydrogen bonds, and the creation of van der Waals forces [206]. Synthetic zeolites (molecular sieves), activated carbon, silica gel, and activated alumina are the most commonly used adsorbents. Physical substances are rapidly absorbed and desorbed, and the process is reversible [207]. However, the rate of chemical adsorption or desorption is slower than that of physical adsorption or desorption because a certain amount of activation energy is required to produce chemical bonds during the chemical adsorption process. Through electrostatic attraction, the ions of the adsorbate concentrate on the charged parts of the adsorbent surface and displace other ions previously anchored in these charged areas. This process is called ion exchange adsorption [208].

Traditional adsorbents have a number of drawbacks, including unpredictable metal ion removal and prohibitively high sludge treatment costs [209]. The need for novel adsorbents with superior adsorption capabilities for minuscule or trace quantities of these contaminants cannot be overstated. These adsorbents must be reasonably priced, user-friendly, and distinctive. Adsorbent design also affects capacity and adsorption rate.

Mesoporous materials exhibit organized pore architectures, high Bragg diffraction peaks in small-angle X-ray diffraction, a narrow pore size distribution, a large specific surface area

(up to 1000 m²/g), and superior thermal stability compared to microscopic and macroscopic materials. Stability (by improving the synthesis process or post-processing) [210]. Mesoporous materials also have a significant amount of unsaturated groups on their surface. Mesoporous materials for catalysis, separations, selective adsorption, and innovative functional materials have seen a sharp rise in attention due to these features [211].

Graphene is thought of as a semimetal because it is a zero-bandgap material with contact between its conduction and valence bands at the Dirac point [212]. It serves as a fundamental building component for many distinct carbon allotropes, including fullerenes, carbon nanotubes, and graphite. In contrast, because of the strong interlayer stacking, hydrophobic contacts, and van der Waals forces between two graphene sheets, graphene compounds quickly cluster and are difficult to disperse in conventional solvents [213]. Consequently, in real-world applications, graphene modification is commonly needed. Several organic-inorganic ordered mesoporous hybrid materials have been designed to filter out impurities (heavy metals, organic contaminants, and nutrients) [214]. Additionally, the industry has made extensive use of graphene oxide composites, an intriguing two-dimensional material with superior physical and chemical capabilities. Mesoporous silica materials structured with graphene oxide may be able to successfully remove heavy metals from wastewater. As, Cd, Cr, Hg, and Pb were all removed at rates of 97.7%, 96.9%, 96.0%, 98.5%, and 78.7%, respectively [215]. This clearly shows how well the material can adsorb contaminants. Materials made by Han and others. It is capable of eliminating phosphates from water and is made utilizing a straightforward one-step sol-gel process [215]. Both in pure and composite settings, GBM demonstrated effective VOC elimination. The most popular graphene structures for removing VOCs are r/GO and GO, which are coating-based [110]. In general, the area for molecular interaction rises along with an adsorbent's surface area. Graphene materials used in composite constructions have a larger surface area than the material's initial GO and rGO forms. Of the materials analyzed, the mixture of GO and MIL-101 had the highest BET surface area, measuring 3502.2 m²/g as opposed to 236.4 m²/g for GO alone.

In general, surface modification can greatly enhance GBM's capacity to absorb VOCs. For instance, r/GO treated with KOH enhanced its ability to adsorb acetaldehyde by 1.23 mg/g. GO surfaces, for instance, are frequently selected to interact with polar VOCs. The interaction with polar VOCs is made simple by the polar functional groups on the surface of GO [216]. On the other hand, hydrophobic contaminants interact more favorably with pure graphene. Instead, r/GO makes distinct choices for polar or nonpolar targets based on the level of GO degradation. Therefore, it is believed that GBM may contribute to the effective removal of VOCs [217].

Graphene oxide (GO), a member of the family of materials made from graphene, is used as the main starting material for the production of graphene. The mechanical, thermal, and electrical properties of this single-layer, spatial, two-dimensional nanomaterial are outstanding. A hexagonal

lattice formed by the bonds between sp² carbons makes up the layered structure of GO [218]. This design's large surface area increases the material's ability to absorb moisture. Every carbon atom is perpendicular to the sheet orbit, which lends weight to the idea that there is an electron cloud. More interactions with adsorbed analytes are possible thanks to Van der Waals forces and hydrogen bonds produced by functional groups with oxygen. Since the surface functional groups may include more surface functional groups and contain oxygen, the material also exhibits hydrophilic characteristics [219]. Both MNP and GO have drawbacks while having good adsorption capabilities. For instance, the active surface of pure inorganic MNPs will decrease when aggregates develop [220]. The magnetic qualities will be lost when Fe₃O₄ (magnetite) with a much decreased magnetic capacity is oxidized into magnetic materials. Fe₂O₃ (maghemite), as well as the laborious, drawn-out, and volumetric SPE process of GO. The GO-MNP linkage addresses these problems. Since the nanoparticles are dispersed on the GO, there is no mixing and the MNPs are not oxidized by ambient oxygen [220]. Due to magnetic separation, the extraction process is faster and less time-consuming than when using GO alone, so no filtration or centrifugation is required. Magnetic graphene oxide (MGO), the final by-product of GO-MNP coupling, has shown remarkable performance as a magnetic adsorbent in MSPE [221].

3.10. Gas sensors

Our daily lives are threatened when we are exposed to hazardous gases in commercial and residential environments [222]. To address health issues, we have developed low-cost, high-performance gas sensors with low-concentration harmful gas detection capabilities. The use of gas sensors is crucial in many fields such as agriculture, medicine, electronics, aviation, etc [223]. The resulting sensor is one of the most intensively studied gas sensors. They do have limitations, however, such as high operating temperatures and long-term stability. Nanotechnology can be used to produce next-generation gas sensing layers with increased sensitivity, selectivity, recovery speed, and reaction time to low gas concentrations [224]. One helpful factor that influences a material's sensitivity is surface area. Due to its inherent electrical characteristics and the large surface area created by its nanostructure, the material known as graphene is regarded to hold potential for sensing applications [225]. It is made up of a sheet of sp²-hybridized carbon atoms that is one atom thick. GO and L-Glu-r/GO's layered and corrugated architectures are advantageous to their ability to detect gases. Higher current and L-Glu-r/Go's thermal stability are two benefits of using L-Glu-r/GO-based sensors at lower voltages (Figure 9) [226]. Although GO is not a new chemical, recent advances in gas sensing technology have focused on its unique characteristics outside of its function as a precursor to reduced graphene oxide (r/GO). While r/GO and graphene are two well-known materials for gas detection, GO is an intriguing material for sensing that has recently garnered a lot of attention.

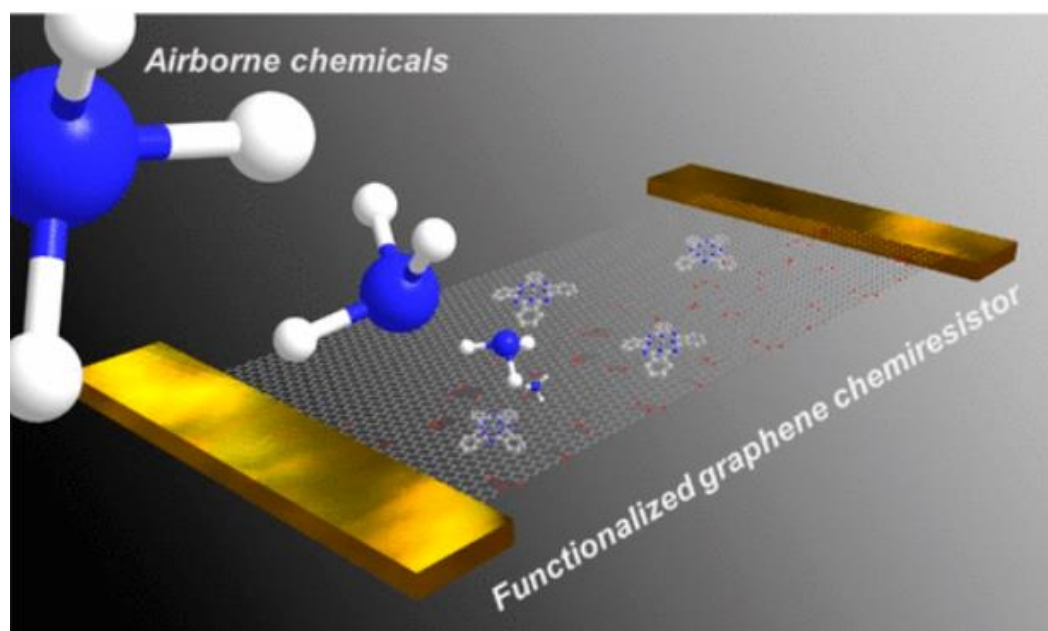


Fig. 9. Sensing application of GO. Reprinted with permission from Ref. [223], Alzate-Carvajal, N., and Luican-Mayer, A., 2020. Functionalized graphene surfaces for selective gas sensing. *ACS Omega*, 5, pp. 21320–21329. Copyright © American Chemical Society.

To make GO materials sensitive and selective to the researched gas, metal nanoparticles and metal oxide nanocomposites can be used [226]. Using graphene as a sensor material for aqueous substances and gas molecules is one of its uses. Since graphene and its analogues have very large specific surface areas, gas sensors are a particularly attractive use for these materials. The way that graphene interacts with various gases may be altered by changing its physical characteristics. According to the Scopus, Schedin made the discovery of graphene-based gas sensors in 2007. Schedin revealed how gas molecules that have been adsorbed raise the carrier density of graphene [227]. A first-principles investigation on the adsorption potential of graphene interacts with electron dopants that are paramagnetic compounds like NO_2 [228].

One may identify gases such as acetone, ethanol, methanol, NO_2 , H_2 , NH_3 , and H_2S with GO-based technology [229]. To create gas sensors with the required sensitivity, selectivity, stability, and repeatability, GO properties must be controlled. The degree of functionalization of the oxygen-containing functional groups of the adsorbed analyte gas molecules is the most important factor. The target analyte and signal detecting technique determine the ideal number of functional groups. Gas sensors for the GO platform are reasonably priced and simple to install [230]. The functional groups on GO are essential for gas detection. They make materials proton conductive and improve interactions with gas molecules [231].

3.12. Humidity sensors

Since graphene nanomaterials exhibit excellent electrical properties, such as strong mobility and minimal electrical signal noise, it is used to build many sensors [232]. This is because of its unique ability to operate under ambient conditions. Due to water and its properties, efficient humidity sensing is possible with response times of up to 30 milliseconds. This humidity-based sensor allows for a variety of applications such as contactless user interfaces that can be displayed through whistle-based identification [233]. One of the most well-known uses of GO is humidity sensing. Since GO has higher proton conductivity and hydrophilicity than graphene and r/GO, it is a better material for water vapor detection than typical proton-conducting materials [234].

The most common humidity measurement is relative humidity (RH), which is important in both domestic and commercial settings [235]. Many industrial fields, including transportation, agriculture, pharmaceuticals, semiconductors, consumer electronics, and food packaging, depend on relative humidity detection. Conducting polymers, metal oxides, and more recently nanocomposites materials made of polymers and nanowires/nanorods are the materials most frequently utilized to make relative humidity sensors [236]. Due to its hydrophilicity, which is largely caused by the presence of organic functional groups like oxygen moieties, graphene oxide (GO), a member of this material family, is the ideal option for relative humidity sensors. The high Young's modulus (32 GPa), huge specific surface area ($736.6 \text{ m}^2\text{g}^{-1}$) and adjustable semiconductivity of GO as a molecular sensing material are additional extremely desired characteristics [237].

3.13. Antibacterial activity

The tremendous growth in infectious illnesses brought on by bacteria, which is alarming millions of people worldwide, is one of the greatest potential threats to human health [238]. Commercial antibiotics function in many different methods, such as by inhibiting the formation of cell walls, stopping the creation of proteins by binding to ribosomes and ribonucleic acid (RNA), stopping DNA replication, etc. Bacterial cells produce enzymes that break down the antibiotic, modify the permeability of their membranes to the medication, alter the specific proteins that the drug targets, and use other tactics to continue to resist it [239]. Multidrug-resistant bacteria, which have evolved as a result of the abrupt abuse of antibiotics, are the most challenging issue affecting human health in the twenty-first century [240]. Furthermore, the availability of such drugs has been steadily declining over time, making nanomaterials an attractive alternative to antimicrobial

agents. However, antibiotics can be effectively replaced by such customized nanoparticles. Shape and size, electronic structure, surface properties, and the environment in which bacterial pathogens interact with nanomaterials (e.g., incubation time, concentration, and media) can all negatively impact the antimicrobial activity of nanomaterials [241]. Compared with antibiotics, antibacterial nanoparticles have two advantages: (a) they have a wide range of bacteria-killing mechanisms, such as cell encapsulation, oxidative stress response, cell membrane rupture, and blocking the entry of nutrients; (b) they can prevent Bacteria grow on solid surfaces (Figure 10) [242]. Oxygen-containing activities adorn the edges and base surfaces of the two-dimensional honeycomb structure of GO. Despite the fact that hydroxyl and carboxyl groups make up the bulk of its basal plane, they also mainly make up its borders. GO interacts more readily with biomolecules including proteins, lipids, and DNA because of its functionality [243].

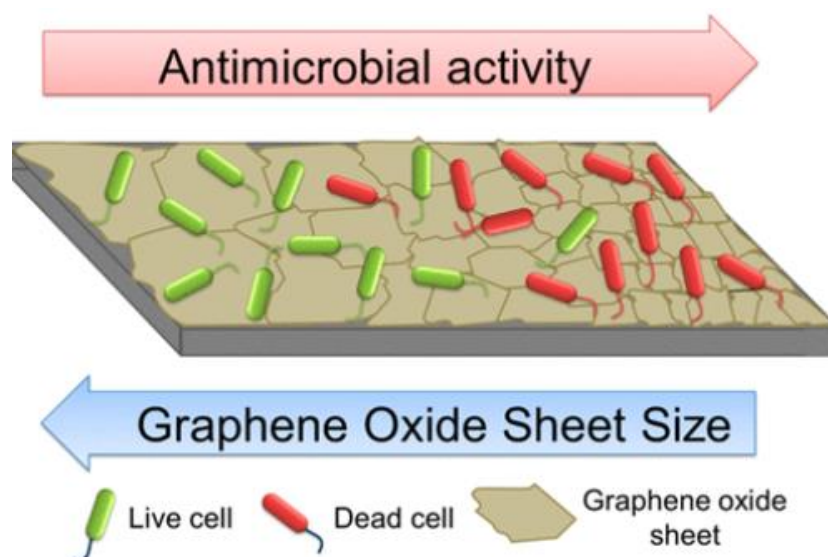


Fig. 10. Antibacterial activity of GO. Reprinted with permission from ref. [243], Perreault, F., de Faria, A. F., Nejati, S., and Elimelech, M., 2015. Antimicrobial properties of graphene oxide nanosheets: Why size matters. *ACS Nano*, 9(7), pp. 7226–7236. Copyright © American Chemical Society.

Graphene or GO, according to several published research, either had no impact on bacteria or encouraged the development of *E. coli* [244]. According to certain reports, materials that resemble graphene can stop Gram-positive bacteria from dividing but not Gram-negative bacteria. Many distinct mechanisms have been hypothesized by various research to explain the bactericidal routes of GO and r/GO. One of the most used techniques is the mechanical rupture of bacterial cells. Physical interactions that GO and r/GO can cause include cell wall breakdown, cell trapping, and cell encapsulation [245]. To stop nutrients from penetrating into the inside of cells and ultimately leading to cell analysis, cells are encircled by a two-dimensional nanomaterial that resembles graphene. When reacts with it, the cell membrane

GO is harmed. A sign of this disruption is cytoplasmic material leakage and structural alterations in the bacterial cell structure. The GO and r/GO's jagged edges create membrane tension, which harms the bacterium physically and eventually tears the cell membrane, enabling RNA to escape.

Redox interactions with biomolecules and the generation of reactive oxygen species (ROS) are also involved in two other bactericidal processes. When biomolecules in bacteria interact with moieties in GO, the redox pathways required for bacterial growth and metabolism are disrupted. When GO interacts with bacterial membranes, carbon free radicals are generated on its surface, which promotes lipid peroxidation by damaging the cell structure, ultimately leading to bacterial cell collapse. Oxidative stress

is a well-established mechanism of interaction between nanoparticles and bacteria [246]. The rupture of bacterial cell membranes is accelerated by the production of lipid peroxides, which are produced when fatty acids in bacteria are oxidized. The cells then lyse and produce small amounts of ROS. According to a recent study, when GO is exposed to *Pseudomonas aeruginosa* cells, it increases the production of ROS, one of the main causes of bacterial cell death [247]. Furthermore, rGO edge defects were found to generate active ROS sites. Membrane rupture occurs when nanoparticles adhere to cell membranes through electrostatic forces. This results in membrane potential alteration, depolarization and damage, leading to transport imbalance, respiratory arrest, and cell lysis and cell death [248].

The suggested technique provides a practical route to create new silver-based antimicrobial compounds [249]. One of the main factors hindering wound healing is local bacterial infection. Many researchers are interested in wound dressings made of antimicrobial materials with added antibiotic drugs. Excellent antimicrobial wound dressings speed up healing by removing harmful bacteria from the wound and maintaining a sterile environment for the healing process [250].

Due to their strong antibacterial properties, CS- and GO-based nanocomposites are often used to heal wounds. The CS:GO nanocomposites wound dressing also exhibited acceptable mechanical strength, anti-inflammatory properties, and tissue adhesion qualities. Furthermore, they are mainly used to fabricate thin films, hydrogels, and nanofibrous membranes [251].

3.14. Anti-viral activity

Numerous studies have been conducted on the ability of graphene-based materials to kill microorganisms. GO and rGO-SO₃ compete with one another for HSV-1's attention because they both contain many negatively charged groups, similar to the cell surface receptor heparan sulfate. The blocking of the virus-binding site by the nanomaterials was the primary inhibitory mechanism that inhibited infection in Vero cells. It is thought that the negatively charged, horny form of GO is what gives it its antiviral abilities [252]. GO conjugated to the nonionic polymer polyvinylpyrrolidone (PVP) showed strong antiviral activity, but no viral inhibition was observed for GO conjugated to the cationic polymer PDDA, demonstrating that negative charge is required for antiviral activity.

Anti-RNA and DNA viruses, enclosed and non-enveloped viruses, sense and negative sense viruses, etc. are all included in the overall antiviral properties of GO and its derivatives. These substances hold significant potential for the creation of antiviral surfaces and coatings that might halt the spread of contaminating agents and viruses like SARS-CoV-2 as shown in Figure 11. Composite structures of GO/rGO-SO₃ with antimicrobial metals such as silver, titanium, and gold should also be investigated to understand more about its potential to develop extremely effective antiviral coating designs. By successfully capturing and crushing the viral structure, these chemicals can shorten the time the virus lives on various coated surfaces [253].

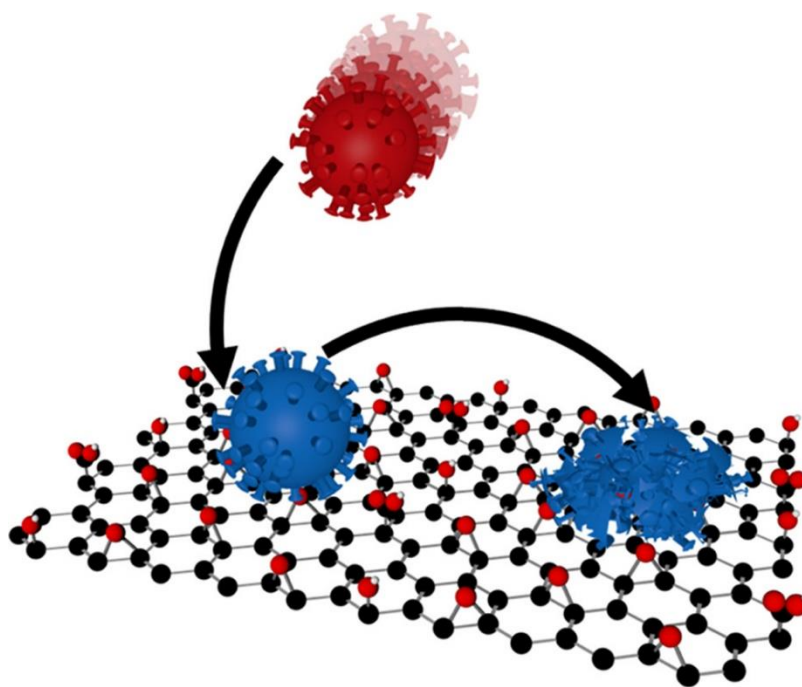


Fig. 11. Adsorption of SARS-CoV-2 by the oxygen-containing functional groups of GO, followed by the reduction of S and N proteins on the GO surface, leading to the inactivation of the SARS-CoV-2 virus. Reprinted with permission from Ref. [253], Fukuda, M., Islam, M.S., Shimizu, R., Nasser, H., Rabin, N.N., Takahashi, Y., Sekine, Y., Lindoy, L.F., Fukuda, T., Ikeda, T. and Hayami, S., 2021. Lethal interactions of SARS-CoV-2 with graphene oxide: Implications for COVID-19 treatment. *ACS Applied Nano Materials*, 4(11), pp.11881-11887. Copyright © American Chemical Society.

A brand-new, fatal SAR-CoV-2 virus was discovered in humans for the first time in December 2019 [254]. Respiratory droplets less than 1 mm in size are the primary method of COVID-19 transmission. After contacting an infected object or surface, a person can get the virus by touching their lips, nose, or eyes. The SAR-CoV-2 virus has variable levels of stability on different surfaces, according to a recent study. SARS-CoV-2 can live on surfaces made of plastic for 72 hours and stainless steel for 48 hours, compared to 4 hours on copper and 24 hours on cardboard [255]. The virus was also proven to be more stable on smooth surfaces than on rough ones, such as wood for two hours, textiles for two hours, and print or tissue paper for three hours. Therefore, infected high-touch surfaces with high viral stability may increase the likelihood of COVID-19 transmission [256].

3.15. Gas transport

The large aspect ratio nanosheet structure of GO-based membranes (high impermeability to most gases) and their negatively charged surface make them a popular choice for gas barrier nanocomposites [257]. Countless instances of fillers like graphene derivatives enhancing barrier characteristics may be found in several publications on polymer nanocomposites. It is understood that 2D fillers may be included into polymer matrices to provide gas molecules a "tortuous" journey, comparable to prior studies of the high gas barrier features of polymer nanocomposites [257]. By employing GO and r/GO nanosheets, the high aspect ratio of the fillers and the oxygen group surface functionalization may be used. According to the green line ("PVA/GO (Small)"), the filler should have a transverse sheet diameter of 0.53 μm and a transverse sheet size of 3.5 μm . The use of this document has been approved by the University of Connecticut. For clarification on how to comprehend allusions to colors in this legend, the reader is recommended to examine the online version of this page [6].

In gas separation operations, GO is frequently disregarded despite its exceptional capacity to generate high barrier membranes for the vast majority of tiny molecular weight species. But in a recent work, scientists showed how intriguing it is to use GO-based membranes to make atomically thin, extremely selective gas transport membranes with specific structural faults that enable the passage of tiny molecules [258, 259]. Gas separation membranes were created to allow for highly selective gas diffusion. The researchers created very selective membranes by fabricating two distinct types of ultrathin GO membranes on polyethersulfone (PES) substrates with thicknesses ranging from 3 to 10 nanometers [260]. By adjusting the gas flow via the GO films' pores and channels, the scientists were able to create these films. In method 1, a GO coating was spin-coated onto the PES surface and it was exposed to the GO solution at the liquid-air interface [261]. GO sheets were piled thicker because of the capillary connections between their faces. The PES surface was immediately spin-coated with the GO solution to achieve this. TEM pictures of GO membrane

cross-sections shows the varied stacking topologies applied for each coating technique. The behavior of membranes created using Procedure 1 may be described by the Knudsen diffusion of gases in nonporous membranes, where the permeability of the membranes decreases with increasing gas molecular weight. Contrary to popular belief, CO_2 does not compare well to other gas concentrations in terms of gas permeability. The permeability of the other gases did not decrease as quickly during the first hour of the permeability test, but that of carbon dioxide did until it leveled out. Using method 1, a high H_2/CO_2 selectivity of 30 was attained. In both dry and wet environments, the interlocking layer structures created by technique 2 react entirely differently from method 1 with CO_2 exhibiting the highest permeability. This technique permits extraordinarily selective diffusion across GO interlayers, displaying improved performance over previously documented polymer membranes since all gases, with the exception of CO_2 , exhibit decreased permeability in wet conditions [262].

3.16. Corrosion resistance

The decomposition of a substance due to its interaction with the environment is called corrosion [263]. It is a global problem with a significant impact on the global economy, with annual losses estimated at \$2.5 trillion, or 3.4% of global GDP. Corrosion can take many different forms, including erosive corrosion, galvanic corrosion, intergranular corrosion, uniform corrosion, pit growth and crevice formation. Many industrial processes, such as pickling, acidifying oil wells, etc., will stimulate direct contact between strong acids and metal surfaces, severely corroding metals and alloys. Many industrial applications, such as cooling systems, boilers, heat exchangers, water treatment facilities, etc., use metallic materials that are attacked by saltwater environments containing chloride ions [264]. Anti-corrosion coatings and corrosion inhibitors are the two most used corrosion protection techniques. Potential ingredients for anticorrosive coatings have included a wide variety of polymers, nanoparticles, and inorganic/organic compounds.

A two-dimensional substance called graphene (G) is formed of Sp^2 hybridized carbon atoms that are arranged hexagonally. G has special chemical properties, thermal stability, and impermeability properties that make it an excellent choice for use as a barrier material in anti-corrosion coatings. On the other hand, graphene oxide (GO), which has functional groups including oxygen, is hydrophilic and has a high water dispersibility [265]. The GO molecule's surface could also make it simpler to functionalize it, which would enhance its performance in terms of dispersibility and corrosion inhibition [266].

Graphene derivatives have the potential to be employed in anti-corrosion polymer coatings due to their exceptional chemical inertness and two-dimensional shape. Fillers and derivatives of graphene can be used to stop these substances from reaching the metal surface and even to make it easier to form a passivation layer. Chemicals that can harm the base

metal, such as chlorides, sulfites, water, and other substances, can only pass through the polymer [266]. Polyaniline (PANI) nanorods, polymerized on nitrogen and fluorine dual-doped graphene oxide (NFGO), enhance the anti-corrosion performance of waterborne epoxy coatings [82]. The PANI/NFGO composites offer high conductivity, improved dispersibility, and effective corrosion resistance, confirmed by electrochemical impedance spectroscopy (EIS) and salt spray tests (Figure 12).

The properties of GO and r/GO in epoxy nanocomposites on steel substrates by modifying the Hummer method and hydrazine reduction, respectively [267]. After 24 hours of exposure to the Cl ion electrolyte flow, electrical impedance spectroscopy (EIS) studies revealed impedance moduli ($|Z|$) of 6.36 k cm^2 and 3.48 k cm^2 for the GO coating and the rGO coating, respectively. Due to its hydrophobicity and tighter interlayer spacing, r/GO demonstrated enhanced electrolyte flow resistance after an hour [268]. But since rGO is better than GO in facilitating the movement of Cl and Fe_3^+ ions through the coating and at the metal/coating contact, it could speed up the generation of corrosion products. According to studies, GO offers steel substrates longer-lasting corrosion protection than r/GO. The corrosion current density of GO (4.86 A cm^2) was much lower after 24 hours of exposure than that of rGO (28.60 A cm^2), and potentiodynamic polarization was observed. Tafel fitting demonstrated that GO-coated steel did not develop any corrosion products [269].

GO-based anti-corrosion coatings have showed promise, but additional study is required to develop corrosion barriers that are reliable and error-free [270]. They contend that to prevent galvanic corrosion, which has proven to be a significant issue, the graphene coating must be completely free of defects and pinholes, as Singhbabu's example demonstrates. Because graphene is a cathodic substance, whether it contains one layer or many, corrosion cannot be prevented once it has started. Even though this issue still

persists for metal anticorrosion coatings, graphene-polymer nanocomposites with properly dispersed graphene and scratch and/or self-healing capabilities may be able to offer a solution [6].

3.17. Energy storage

Because of their superior electrical conductivity, great mechanical strength, significant surface area, and superior chemical stability, materials based on graphene have drawn significant interest in the field of energy storage [271]. In energy storage devices like supercapacitors and batteries, it is much simpler to employ 3D printed graphene with higher mass loading and other programmable properties. Supercapacitors are recognized as significant energy storage technologies because of their high power density, low price, and lengthy charge and discharge lifetimes [272]. Through charge separation, they store energy at the electrochemical contact between electrodes and electrolytes. The operational efficiency of supercapacitors is significantly influenced by the material qualities of the electrode and electrolyte. Three-dimensional graphene structures fabricated via 3D printing are promising and frequently demonstrated (Figure 13).

Stretchable electronics, lithium-ion batteries (LIBs), and supercapacitors are just a few of the energy-related applications where rGO is a potential material due to its superior electrical properties [273]. By mixing GO with PU sponge template GO supercapacitor electrodes were created. Due to its 3D structure and internal porosity, the material has an energy density of 89 W h kg^{-1} in aqueous solution and a specific capacitance of 401 F g^{-1} [6].

Due to its high power, cycle stability, high energy density, expanded surface area, and cheap cost, supercapacitors, also known as electrochemical capacitors (EC) or supercapacitors (SC), have gained importance as a research topic.

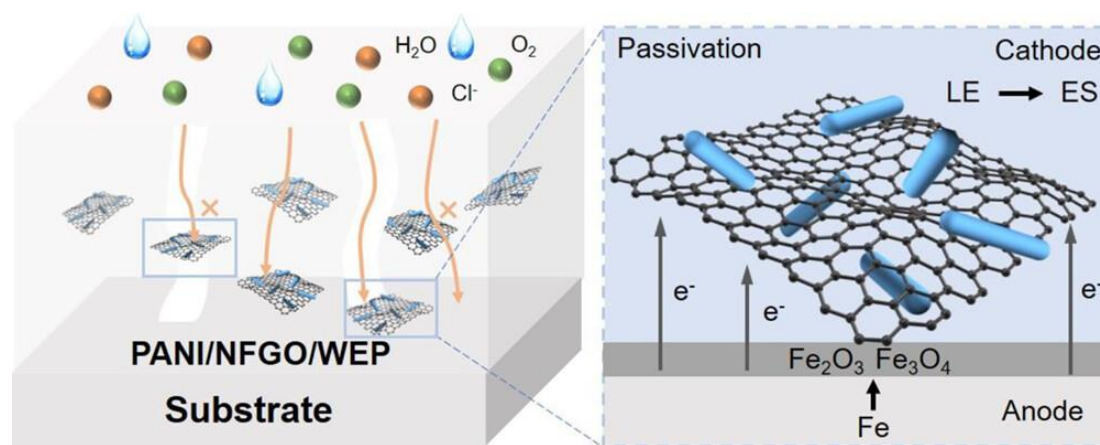


Fig. 12. Schematic of the corrosion protection mechanism. Reprinted with Permission from Ref. [82], An, H., Liu, K., Wang, S., Jiang, C., Liang, S., Xiao, J., Zhao, X., Sun, Z., Cao, C. and Gao, Y., 2023. Enhanced corrosion resistance of waterborne epoxy coatings by polyaniline nanorods and nitrogen and fluorine dual-doped graphene oxide composites. *ACS Applied Nano Materials*, 6(14), pp.13250-13259. Copyright © American Chemical Society.

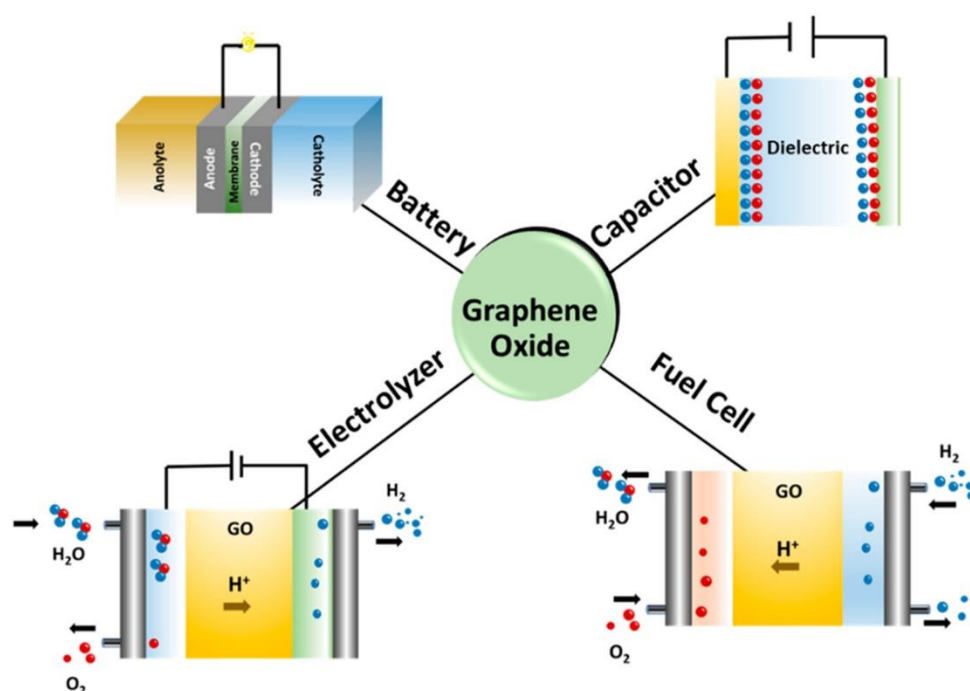


Fig. 13. Schematic representation of different energy applications of GO. Reprinted with permission from ref. [273], Gautam, M., Kanade, S., and Kale, B. B., 2023. Electrochemical energy storage and conversion applications of graphene oxide: A review. *Energy & Fuels*, 37(22), pp. 17134-17160. Copyright © American Chemical Society.

Graphene, a two-dimensional sp^2 -bonded carbon atom, has recently attracted a lot of interest due to its extraordinary properties, including high electrical conductivity, thermal conductivity, mechanical strength, specific surface area, and potentially lower production costs. This fundamental idea underlies the operation of capacitor energy storage, which accounts for its extensive use [274].

Despite being a non-metal, graphene is frequently referred to as a metalloid because to its physical similarities to semiconductor metals. Each carbon atom has a free electron and is covalently bonded to three other carbon atoms in the hexagonal configuration (sp^2 hybridization). The unbound electrons are found in the p orbital, which is located above the plane of matter. Due to the existence of two delocalized electrons in each hexagon, graphene sheets have a high electrical conductivity [275].

Its great electrical conductivity is graphene's most pervasive and important property [276]. No electronic band gap exists in graphene, hence it cannot be extended or closed. This is due to the fact that electrons behave as massless relativistic particles and the valence and conduction bands seldom ever overlap. Due to its homogenous surface area, consistent pore size distribution, and superior electrolytic surface exposure, graphene is regarded as a promising supercapacitor component [276].

High-performance supercapacitors use functionalized nanostructured carbon materials as its primary electrode component [277]. Three-dimensional (3-D) graphene materials and heteroatom doping have special benefits that distinguish them from other arrangements. Numerous active

sites for electrochemical reactions are provided by the heteroatom-doped structure, and the three-dimensional nature of the structure is advantageous for ion transport.

Three-dimensional graphene is the perfect three-dimensional electrode material for supercapacitors because it can primarily prevent the irreversible stacking effect of two-dimensional nanosheets. The distinctive 3D architecture of graphene facilitates effective charge and ion movement to the active material, enhancing its capacity for charge storage and producing high energy and power densities [278].

The porous graphene structure has incredible mechanical strength, allowing it to accommodate significant volume changes and reduce mechanical stress, enabling long-term cycling stability. There is considerable interest in doping carbon materials at the atomic level to achieve variable properties. Heteroatom (N, S, B, P, etc.) doping can successfully enhance the electrical and chemical reactivity of graphene, thereby greatly improving the electrochemical performance of the material [279]. Heteroatom-doped graphene materials with altered electronic structures can provide unusual electrochemical active sites with small changes in conjugation length due to significant charge polarization. This is based on the different electro negativities of heteroatoms and carbon atoms.

Furthermore, by reducing the inertia of the graphene layer, doping heteroatoms in the graphene skeleton with a certain atomic radius can increase the degree of disorder, hence enhancing the specific capacitance and power quality [280]. S and P, two heteroatoms, are among them. Thirdly, the van der Waals force between graphene nanosheets and the

chemical reactivity of graphene are modified by certain heteroatoms' lone pairs of electrons, which serve as carriers to promote electron migration. Use of graphene, which frequently has significant surface activity, may considerably improve the performance and pseudocapacitance of supercapacitors [281-288].

The usage of r/GO-based materials in lithium-ion batteries has proven successful in various energy storage applications [283]. It was employed as an electrode for LIB. When compared to lithium batteries, sodium-ion batteries (SIBs) are more affordable and environmentally friendly [116]. Graphene oxide and CuCO_2S_4 nanocomposites were used as anode materials for a sodium-ion battery prepared by a one-step solvothermal technique. The material was examined by several techniques. In terms of storage effectiveness and electrochemical performance, the nanocomposite, according to this theory [289-295]. Therefore, graphene-based nanocomposites can be used as anode materials in battery structures [296].

3.18. Solar cells

Over the years, the development of new solar power generation, storage and utilization technologies has stimulated interest in finding alternative strategies for clean energy [299-301]. Sunlight is a plentiful, secure, economical, and eco-friendly energy source since it can be transformed directly into electricity without producing pollution or other environmental issues [302-304]. Photovoltaic materials and devices assist the photoelectric effect, converting light energy into electrical energy. Graphene is a game-changer for conductive transparent electronics and presents special

prospects to the energy industry because of its amazing two-dimensional (2D) structure. Graphene has been used in solar cell systems for the past 20 years as a transparent electrode, a hole/electron transport medium, and an interfacial buffer layer [306-308]. It is an electrical device that generates energy by irradiating solar cells with sunlight to generate voltage and current. As a result, electricity is generated. Transparent graphene and similar nanomaterial cathode electrodes are also used in solar cells, according to the study. They used reduced graphene oxide as the working electrode and cathode, enabling the electrochemical deposition of materials in solar cells. Due to graphene's electrical properties, it can be used in solar cell-based applications [309].

Zannotti et al. reported the synthesis of reduced graphene, and NiO/rGO [310]. The reduced graphene oxide (rGO) was prepared from commercial graphene oxide using a thermal reduction method, while the NiO/rGO photocathodes were prepared through a mixed NiCl_2/rGO sol-gel process with varying rGO amounts. Detailed characterization using SEM, Raman, and BET analysis revealed optimal surface areas ranging from 0.0 to 0.3 mg of rGO. Higher rGO content, despite lower surface area, facilitated increased dye uptake due to π -stacking interactions between dye phenyl groups and graphene sheets. The p-type dye-sensitized solar cells (DSSCs) made from these films exhibited a significant increase in short-circuit current density (JSC) up to 2.6 mA/cm^2 and an open-circuit voltage (VOC) of 125 mV, doubling the efficiency compared to NiO-only cells. This enhancement is attributed to higher dye uptake, leading to increased photoexcitation and hole concentration in the photocathodes (Figure 14).

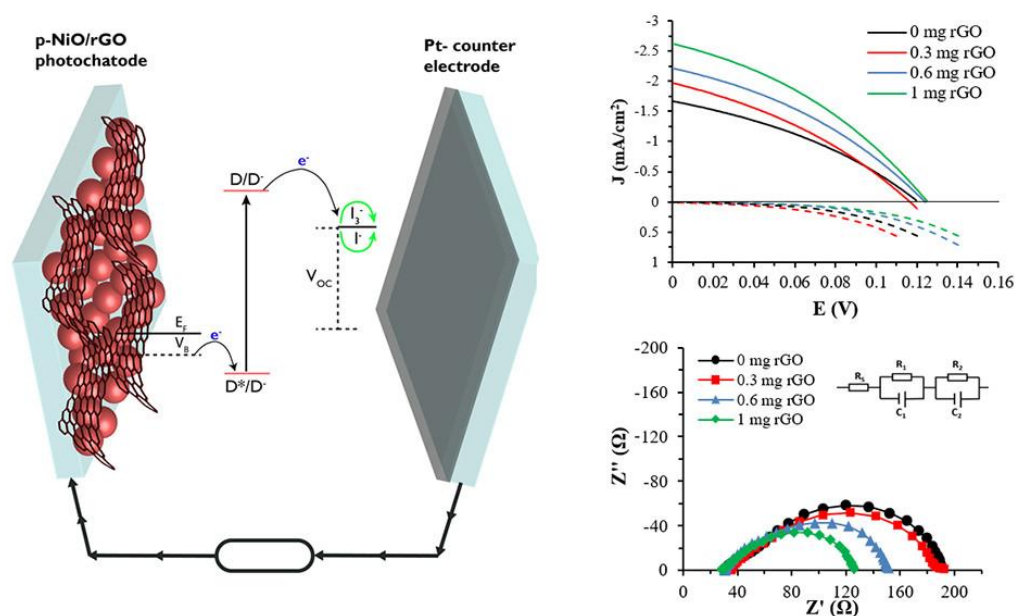


Fig. 14. Reduced graphene oxide-NiO photocathodes for p-type dye-sensitized solar cells. Reprinted with permission from Ref. [310], Zannotti, M., Benazzi, E., Stevens, L.A., Minicucci, M., Bruce, L., Snape, C.E., Gibson, E.A. and Giovannetti, R., 2019. Reduced graphene oxide-NiO photocathodes for p-type dye-sensitized solar cells. *ACS Applied Energy Materials*, 2(10), pp.7345-7353. Copyright © American Chemical Society.

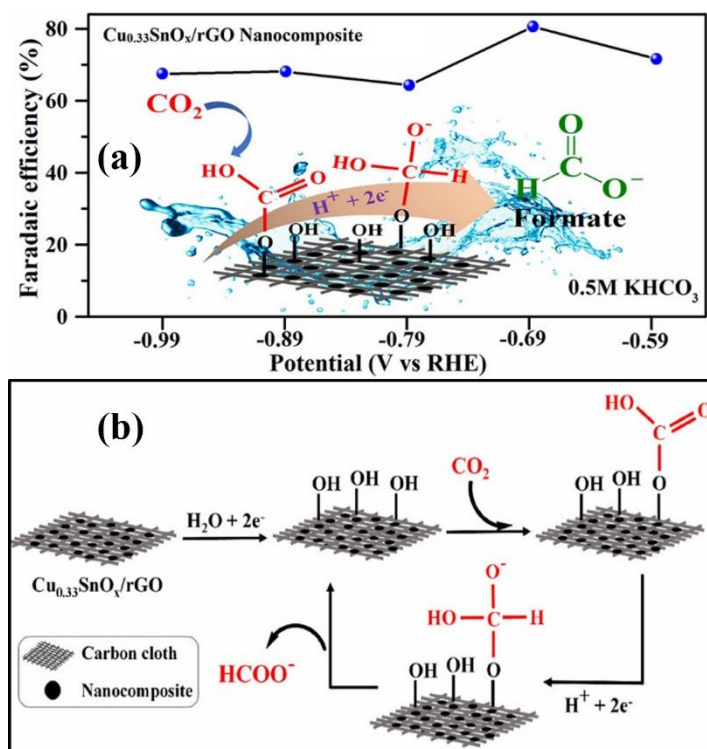


Fig. 15. Electrochemical CO_2 reduction over rGO. Reprinted with permission from ref. [312], Kempasiddaiah, M., Samanta, R., Panigrahy, S. and Barman, S., 2023. Interface-rich highly oxophilic copper/tin–oxide nanocomposite on reduced graphene oxide for efficient electroreduction of CO_2 to formate. *ACS Applied Energy Materials*, 6(5), pp.3020-3031. Copyright © American Chemical Society.

3.19. Electrochemical CO_2 reduction

Upstream energy demand will be four times the present world consumption by 2050 as a result of the growth of human society and the industrial economy [311, 312]. Fossil fuels are expected to be the dominant source of energy for many years to come. Due to their use and the resulting large emissions of carbon dioxide, they contribute to serious resource, environmental and climate problems (also known as "global warming"). An early solution to preventing these catastrophic effects and reducing atmospheric carbon dioxide concentrations appears to be direct electrochemical carbon dioxide reduction (ECR), which produces fuel and common chemicals (oxidized hydrocarbons) [313].

By producing power from renewable energy sources (solar, tidal, wind, and geothermal energy) at the anticipated low rates, it is possible to achieve the energy transition from a "fossil fuel economy" to a sustainable " CO_2 economy." The reaction between waste carbon dioxide and water is propelled by this energy. Catalysts for CO_2 electroreduction might be homogeneous or heterogeneous. Because each active site can be precisely controlled, homogeneous catalytic systems, including enzymes and molecular catalysts, have benefits in terms of effectiveness and product selectivity. However, because of their expensive price, poor stability, and challenging post-stage separation, they are constrained in their scalability and applicability. The solution to these

problems, which necessitates immobilization on a conductive support like graphene, has just lately been discovered [314].

Recent research shows bimetallic electrocatalysts improve electrochemical formate production during CO_2 reduction but have issues with selectivity and durability [312]. This study enhances CO_2 reduction to formate by incorporating reduced graphene oxide (rGO) into nanostructured bimetallic CuSnO_x electrocatalysts ($\text{Cu}_x\text{SnO}_x/\text{rGO}$). The interface-rich, mixed crystalline–amorphous $\text{Cu}_{0.33}\text{SnO}_x/\text{rGO}$ nanocomposite lowers the over potential to 590 mV vs RHE. SnO_x increases electrocatalytic activity and selectivity for formate. The nanocomposite achieves an 80.62% faradaic efficiency at -0.69 V (RHE), outperforming $\text{CuSnO}_x/\text{rGO}$ and $\text{Cu}_3\text{SnO}_x/\text{rGO}$. The improved performance is due to synergistic interactions, interfaces, higher electrochemical surface area, and its mixed crystalline–amorphous structure (Figure 15).

The research on chalcogenides, carbides, carbon-based materials and metal-organic framework materials is of great significance. Transition Bimetals (such as Pt, Rh, Ni, Pd, Ag, Au and Cu) and p-block Bimetals (such as Pb, Sn, In and Bi) and oxides (such as RuO_2 , IrO_2 , TiO_2 and SnO_2) have also received considerable attention. Carbon-based materials stand out among competitors in the ECR industry because of their potential [315]. They possess a variety of beneficial qualities, including abundance, economy, good electrical conductivity, excellent structural stability, environmental

friendliness, and renewable nature. When employed as a catalyst or catalytic support, it also offers exceptional mechanical stability, enormous mechanical strength, and amazing electrical conductivity.

The ease with which metallic and non-metallic compounds may alter the surface of graphene is another crucial factor. This makes it possible to adjust the relative catalytic activity somewhat. Since neutral carbon atoms cannot activate CO₂, both pure graphene and graphene oxide (GO) are inert to electrochemical reactions (ECR). Doping graphene with heteroatoms such as boron, nitrogen, and phosphorus will greatly enhance its ECR activity while also significantly changing the electrical structure of the material (including single, double, and multiple heteroatom doping) [316]. The creation of graphene-based heterostructures can also enhance charge transfer and create beneficial synergistic effects, thereby promoting the reduction of carbon dioxide emissions.

3.20. Use in Lubricants

One of the most useful items for everyday use is lubricants, as they help reduce friction and equipment wear. Therefore, use lubricants to extend the service life of your machine, especially under heavy loads.

Liang et al. explored the potential of graphene oxide (GO) film as a solid lubricant for silicon-based MEMS devices [317]. Using a green electrophoretic deposition (EPD) method, GO films with nanoscale tunable thickness were fabricated on silicon wafers. Characterization of the films revealed a significant reduction in the friction coefficient to 1/6 and a decrease in wear volume to 1/24 of their original values. These results suggest that GO films are excellent solid lubricants. The EPD technique allows control over film thickness by adjusting the working voltage, leading to harder, rougher films with improved adhesion and wear resistance at higher voltages. Overall, GO films significantly enhance friction reduction and wear life, making them promising solid lubricants for MEMS/NEMS devices.

Chouhan et al. present a scalable method to create

aminoborate-functionalized reduced graphene oxide (rGO-AmB) for aqueous lubricants [318]. Characterized by several techniques, rGO-AmB demonstrates multiple interaction pathways with aminoborate (AmB). Notably, rGO-AmB offers long-term dispersion stability and boosts water's thermal conductivity by 68%, with increased performance at higher concentrations and temperatures. When added to water (0.2%), rGO-AmB significantly enhances the tribological properties of a steel tribopair, reducing friction by 70% and wear by 68%. This improvement is due to the gradual deposition of a thin rGO-AmB film, which aids interfacial sliding and protects against wear (Figure 16). rGO-AmB's ultrathin structure, excellent water dispersibility, high thermal conductivity, low friction, and strong affinity for tribo-interfaces highlight its potential as an effective additive for aqueous lubricants, offering a green and efficient solution for enhanced lubrication.

Savjani et al. developed fluorinated graphene oxide–copper metal matrix composites (FGO-CMMC) for enhancing the tribological properties of copper coatings on steel switching mechanisms [319]. These coatings were fabricated on AISI 52100 bearing steel substrates via a simple electrodeposition process in water. The FGO-CMMC coatings demonstrated excellent lubrication performance under pin-on-disk tribological sliding at a 1N load, reducing the coefficient of friction (CoF) by 63% and 69% compared to GO-CMMC and pure copper coatings, respectively. The enhanced lubrication is attributed to the tribochemical reaction of FGO with the steel counterface, leading to the formation of an asymmetric tribofilm structure that passivates the system from adhesion-driven breakdown. Additionally, FGO-CMMC and GO-CMMC coatings significantly improved corrosion protection, with efficiencies of 94.2% and 91.6% compared to the bare steel substrate. The improved interlaminar shear strength and the robust, asymmetrically structured tribofilm contribute to the excellent lubrication and wear resistance. These properties make FGO-CMMC a promising solid lubricant candidate for mechanical engineering applications, particularly in steel-based mechanical switch assemblies.

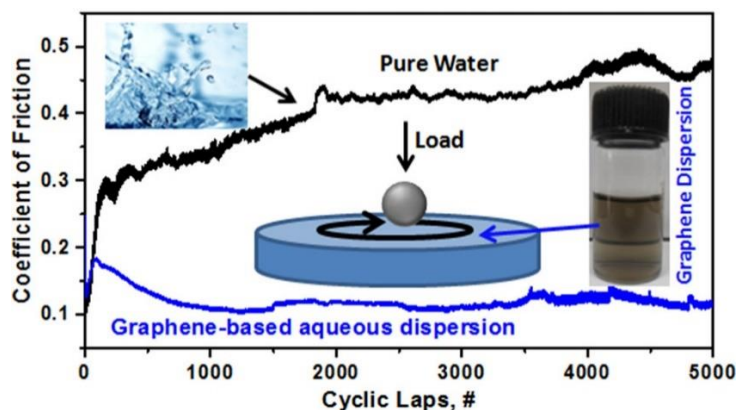


Fig. 16. Friction reduction by aqueous dispersion of chemically derived graphene. Reprinted with permission from ref. [318], Chouhan, A., Kumari, S., Sarkar, T.K., Rawat, S.S. and Khatri, O.P., 2020. Graphene-based aqueous lubricants: Dispersion stability to the enhancement of tribological properties. *ACS Applied Materials & Interfaces*, 12(46), pp.51785-51796. Copyright © American Chemical Society.

3.21. Graphene oxide in agriculture

Graphene and related nanoparticles are gaining popularity in agriculture due to their unique physicochemical properties and their established applications in the electronics and pharmaceutical sectors [320]. Various research teams have developed a graphene oxide and silver nanocomposite (GO-AgNPs) which has shown effectiveness as an antifungal agent against *Fusarium graminearum*, a pathogen responsible for crop diseases such as leaf spot infection. Studies demonstrated that the inhibitory efficiency of GO-AgNPs, with inhibition levels ranging from approximately 3 to 7, surpasses that of pure silver and graphene solutions. The minimum inhibitory concentration (MIC) necessary to inhibit spore germination was found to be 4.68 $\mu\text{g/mL}$. These findings suggest that graphene and graphene-based nanocomposites are promising components for developing new antifungal agents to combat agricultural diseases [320].

Additionally, researchers have developed a phosphate-containing nanocomposite (graphene oxide/iron) aimed at enhancing nutrient delivery to plants [321]. Characterized using various techniques, the GO-Fe composite exhibited a phosphate loading capacity of 48 mg P/g, attributed to the ferric ions' ability to bind phosphate ions. When compared with conventional fertilizers like monoammonium phosphate (MAP), the novel graphene/iron/phosphate nanocomposite demonstrated a slower phosphate release, reducing the risk of soluble phosphate leaching into groundwater. This innovative approach holds promise for improving farm management practices by incorporating advanced materials like graphene [321].

Furthermore, the application of graphene oxide at a concentration of 50 mg/L has been shown to significantly enhance the photosynthetic capacity, yield, nutrient content, and morphological characteristics of plant leaves. In the case of aloe vera, graphene oxide was found to accelerate growth by promoting root development and photosynthesis. GO enhanced biomass accumulation in roots, stems, and shoots by increasing auxin concentration and the expression of genes related to root development [322]. For tomato plants, an appropriate graphene oxide concentration (50-100 mg/L) has been observed to increase yields, demonstrating its potential benefits for agricultural productivity [322].

4. CONCLUSION

Recently, interest in graphene oxide (GO) materials has surged as various industries have increasingly incorporated these compounds into their processes. The production of GO typically follows the methods proposed by Brodie, Staudenmaier, and Tour, with ongoing discussions about techniques to produce GO on a large scale using potential oxidants. Significant advancements have been made in recent years, particularly with the development of numerous new, improved, and modified Hummer's procedures. These procedures have proven to be effective for large-scale GO production, facilitating its use in various applications

including energy storage, water purification, biosensors, electronics, and the elimination of hazardous gases. This work highlights the practical and accessible nature of Hummer's and Modified Hummer's techniques for producing GO on a wider scale, showcasing the fascinating and distinctive properties of synthesized GO. This review serves as a valuable resource for upcoming researchers, offering comprehensive insights into the diverse literature on GO and its potential applications.

5. FUTURE PERSPECTIVES

Future research should focus on optimizing production techniques to enhance the scalability and cost-effectiveness of graphene oxide (GO) synthesis. Investigating alternative, environmentally friendly oxidants and refining existing methods can lead to more efficient production processes. Exploring the functionalization of GO with various chemical groups and the creation of hybrid materials can expand its applications, including developing GO composites with enhanced properties for specific uses in medical, environmental, and industrial fields. Building on the current understanding of GO's role in agriculture, future studies could investigate its impact on a wider range of crops, soil types, and environmental conditions, including exploring its potential as a carrier for fertilizers and pesticides to improve crop yield and health. Further research into the integration of GO in energy storage systems, such as batteries and supercapacitors, can lead to significant advancements in energy efficiency and capacity, with a focus on developing GO-based materials with higher conductivity and stability for next-generation energy storage technologies. Expanding the use of GO in environmental remediation, particularly in water purification and air quality improvement, is another promising area for future research, including investigating the mechanisms of pollutant adsorption and degradation on GO surfaces to lead to more effective solutions for environmental cleanup. Exploring the potential of GO in biomedical applications, such as drug delivery systems, biosensors, and tissue engineering, can open new avenues for medical advancements, with a focus on researching the biocompatibility and targeted delivery mechanisms of GO-based materials to lead to innovative healthcare solutions. As the use of GO expands, it is essential to conduct comprehensive studies on its environmental impact, toxicity, and long-term effects, establishing clear guidelines and safety protocols to ensure the responsible and sustainable use of GO in various industries. By addressing these future directions, researchers can unlock the full potential of graphene oxide, paving the way for groundbreaking advancements across multiple disciplines.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests.

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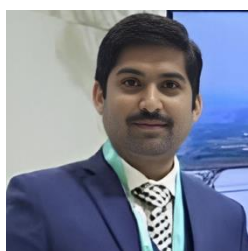
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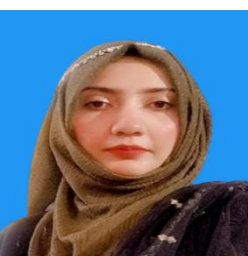
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