

REVIEW

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# Graphene oxide synthesis and applications in emerging contaminant removal: a comprehensive review

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## Abstract

This thorough review explores the pioneering applications of graphene oxide (GO) in tackling emerging environmental pollutants, highlighting its distinct role in environmental remediation. Setting itself apart, this review meticulously synthesizes cutting-edge research, focusing on GO's practical applications in eliminating emerging contaminants from water. It is worth highlighting that there is a limited number of reviews focused on this particular subject, making this work outstanding. It provides specific instances of successful contaminant removal, identifies knowledge gaps, and proposes future directions. Serving as a vital resource for researchers and practitioners, it offers practical insights into applying GO in contaminant remediation, especially in challenging environments. The review critically analyzes crucial gaps in current research, including understanding the long-term environmental effects of GO, its interactions with diverse pollutants, and effective large-scale implementation. This review not only expands our knowledge, but also guides future research endeavors. Furthermore, it outlines clear pathways for future studies, advocating for in-depth ecological research, advanced contaminant interaction analyses, and innovative large-scale implementation strategies. This work establishes a strong foundation, defining the unique novelty of GO applications in environmental remediation and shaping the future discourse in this essential field of study.

**Keywords** Graphene oxide, Synthesis, Efficient removal, Emerging contaminants, Traditional treatment methods, Environmental remediation

## Introduction

Emerging contaminants are substances that have recently been identified as potential threats to human and environmental health, but for which regulatory limits and

treatment methods have not yet been established [1]. These contaminants can come from a variety of sources, including industrial, agricultural, and domestic activities, and can enter water systems through direct discharge or through runoff from land [2]. Examples of emerging contaminants include pharmaceuticals, personal care products, endocrine-disrupting chemicals, and microplastics [3]. These substances can have adverse effects on aquatic organisms and ecosystems, and some have been linked to health problems in humans, including hormone disruption, cancer, and neurological disorders [4].

Emerging contaminants pose a challenge to water treatment due to their persistence, low concentrations, and diverse chemical properties [5]. Traditional treatment methods, such as filtration and chlorination, may not be effective in removing these contaminants [6]. Therefore,

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new technologies and treatment methods are needed to address this growing problem. Graphene oxide is one such technology that has shown promise in the removal of emerging contaminants from water [7].

Graphene-based materials, including graphene oxide, have shown promising results in the remediation of emerging contaminants from water [8]. Graphene oxide has a high surface area, which provides more active sites for chemical reactions, and it also has a strong affinity for organic and inorganic pollutants [9]. One of the key advantages of using graphene oxide in the remediation of emerging contaminants is its ability to remove a wide range of substances, including pharmaceuticals, personal care products, and endocrine-disrupting chemicals, among others [10]. Graphene oxide can also be modified to target specific contaminants, making it a versatile tool for water treatment [11]. Another advantage of using graphene oxide is its low cost and abundance [12]. Graphene oxide can be produced from inexpensive starting materials and can be easily scaled up for industrial use [13]. Furthermore, graphene oxide is a stable material that can withstand harsh treatment conditions, such as high temperature and pressure [14]. This makes it a robust option for use in water treatment systems. In general, the use of graphene oxide in the remediation of emerging contaminants has the potential to provide a cost-effective and efficient solution to this growing problem [15]. As research continues to explore the full capabilities of graphene oxide in water treatment, its use in the removal of emerging contaminants is expected to increase [16].

Graphene oxide has gained increasing attention as a potential material for water treatment due to its unique properties, such as a large surface area, high adsorption capacity, and chemical stability [17]. Graphene oxide can be functionalized to selectively target specific contaminants, making it a versatile tool for water treatment [18]. In the context of water treatment, graphene oxide has been used for the removal of various contaminants, including heavy metals, organic compounds, and microorganisms [19]. Graphene oxide can be used alone or in combination with other materials, such as activated carbon or nanoparticles, to enhance its performance in water treatment [20]. Graphene oxide can also be used for the development of membrane-based technologies for water treatment, such as ultrafiltration and nanofiltration membranes [21]. The incorporation of graphene oxide into these membranes can enhance their performance, such as improved selectivity and anti-fouling properties [22]. The use of graphene oxide in water treatment also offers potential benefits for environmental sustainability. For example, graphene oxide can be derived from renewable resources, and its use in water treatment can reduce the amount of chemical treatment required

for water purification [23]. However, the application of graphene oxide in water treatment is still in the early stages of development, and several challenges need to be addressed, such as the potential release of graphene oxide particles into water systems and the high cost of production [24]. Further research is needed to optimize the use of graphene oxide in water treatment and to assess its long-term environmental impact [16].

This review stands out in its field. While many other reviews only look at the scientific aspects, this study goes deeper. It explores how graphene oxide is made and how it is used practically. Unlike theoretical discussions, it looks at real situations, proving how effective graphene oxide is at removing harmful pollutants like drugs and pesticides. It gives specific examples of successful contaminant removal, giving researchers and practitioners valuable knowledge on how to use this material effectively. Previous studies have confirmed that graphene oxide effectively removes emerging contaminants from water by processes like adsorption and catalytic degradation [14, 15, 25, 26]. However, there is still a significant gap in the literature when it comes to a detailed review focused on using graphene oxide for emerging contaminant cleanup. Such a review not only identifies areas needing more research, but also highlights the difficulties and possibilities in using graphene oxide for this purpose. Furthermore, it outlines the environmental and economic benefits linked with this technology.

As a result, this review seeks to furnish an extensive overview of the properties and applications of graphene oxide in water treatment, with a particular emphasis on its potential for the removal of emerging contaminants.

## **Graphene oxide synthesis and properties**

### **Methods of synthesis**

Graphene oxide (GO) can be synthesized using various methods, each with its advantages and disadvantages. The most commonly used methods for GO synthesis are the Hummers method, Staudenmaier method, and Tour method [27].

### **Synthesis of graphene oxide using Hummers method**

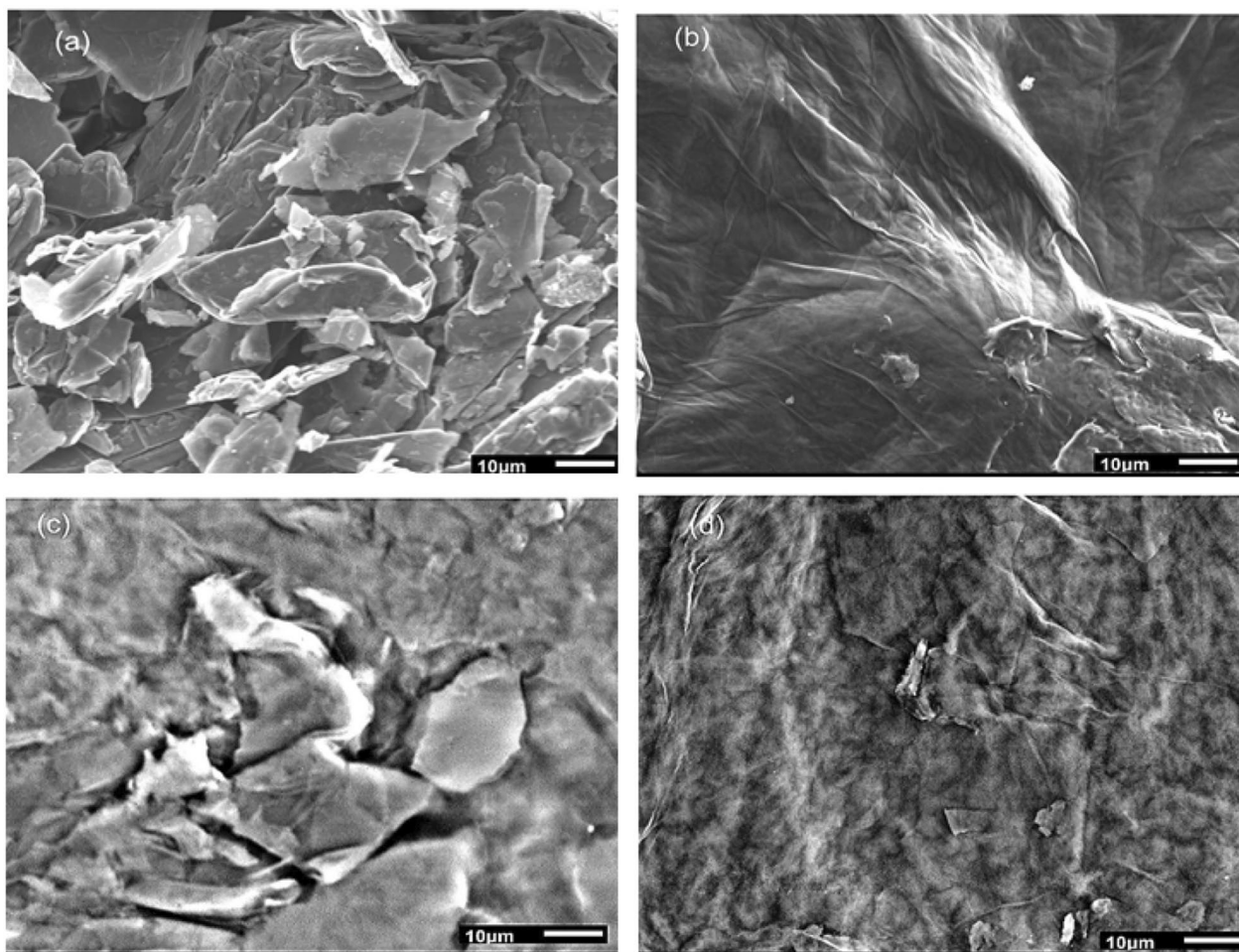
The Hummers method is the most widely used method for GO synthesis. In this method, graphite is oxidized with a mixture of strong acids, such as sulfuric acid and nitric acid, along with potassium permanganate [28]. The resulting graphite oxide is then sonicated and washed with deionized water to remove excess acid and salts. The resulting GO has a high concentration of oxygen functional groups, making it highly hydrophilic [29]. In recent times, there have been several studies on the synthesis and characterization of GO using the Hummers method [30, 31].

Several studies have focused on the synthesis of graphene oxide using the Hummers method, which is a widely used technique for preparing graphene oxide from graphite [32, 33]. Graphene oxide has attracted significant attention due to its unique properties and potential applications in various fields. However, the traditional Hummers method has limitations in terms of efficiency and scalability. In this study, Guo et al. propose a modified approach by introducing an electric field during the synthesis process to enhance the efficiency of graphene oxide production [32]. They investigate the effects of different parameters such as the electric field strength, reaction time, and temperature on the synthesis process. The results demonstrate that the application of an electric field can significantly improve the efficiency of graphene oxide synthesis, leading to higher yields and shorter reaction times compared to the traditional method. The findings of this study provide valuable insights into the synthesis of graphene oxide and offer a potential strategy to enhance the efficiency of the process.

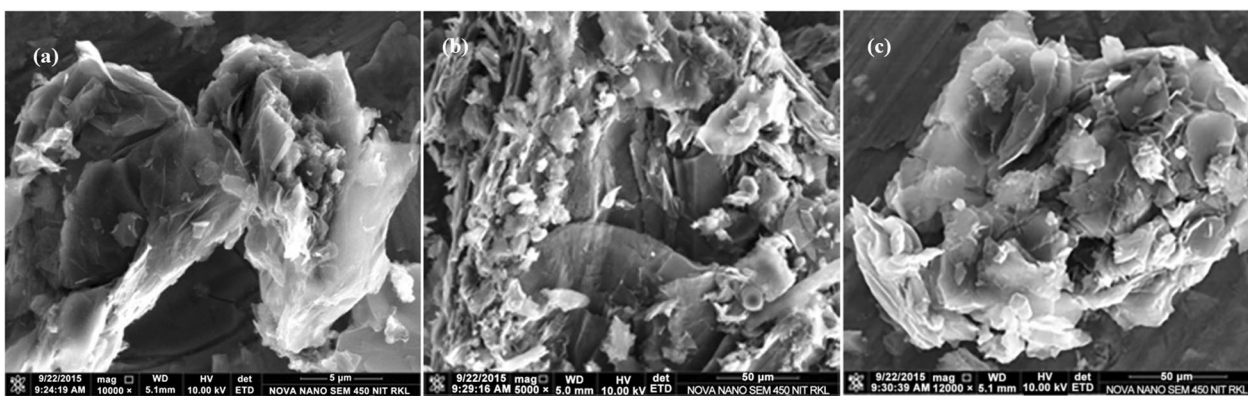
In a study conducted by Olorunkosebi et al. the optimization of graphene oxide (GO) synthesis using different variations of the Hummers' method was investigated [33]. The researchers explored the effects of various reaction parameters, including oxidizing agent concentration, reaction time, and temperature, on GO synthesis. Neem and pumpkin leaf extracts were utilized as reducing agents. The properties of GO samples prepared using different variations of the Hummers' method were compared, and their suitability for reduction to obtain reduced graphene oxide (rGO) was evaluated. Characterization of the obtained GOs and RGOs was performed using Fourier infra-red (FTIR) spectroscopy, Raman spectroscopy, UV–visible spectrophotometry, energy dispersive X-ray spectroscopy (EDX), and scanning electron microscopy (SEM). The results demonstrated that all three methods employed were capable of producing GO with varying degrees of oxidation. Analysis of FTIR spectra of RGOs revealed the absence of the C–O signature observed in GOs at 1730–1740  $\text{cm}^{-1}$  after reduction. SEM micrographs of the graphite and synthesized GOs exhibited a flake-like shape, prominently visible at the edges, with multiple layers arranged on top of each other (see Fig. 1a). For GO12H (GO stirred for 12 h) in Fig. 1b, an ultra-thin layer with folded edges was observed, indicating a continuous surface. Individual sheets could be differentiated through their edges, featuring wrinkled and kinked areas. These sheets were intercalated with rough surfaces resulting from structural deformation during exfoliation and restacking. The morphology of GOSIM (powdered form GO) in Fig. 1d was similar, with slight differences attributed to the level of oxidation. In

the case of GONP in Fig. 1c, a structure at the center resembling graphite in Fig. 1a was observed, possibly due to the level of oxidation. Additionally, other characterization outcomes of the RGOs demonstrated the effectiveness of both neem and pumpkin extracts as reducing agents for the synthesized GO. Consequently, it can be inferred that neem and pumpkin extracts hold promising potential as reducing agents. The findings of this study provide valuable insights into the optimization of graphene oxide synthesis using the Hummers' method and offer a comparative analysis of different reduction approaches.

In a previous investigation, graphene oxide (GO) was prepared using a modified Hummers method, which involved oxidizing purified natural flake graphite (NFG). The SEM images in Fig. 2a–c depict the as-received NFG, illustrating the effects of the oxidation process. This process generated a brown-colored viscous slurry consisting of graphite oxide, exfoliated sheets, non-oxidized graphitic particles, and residual oxidizing agents. The synthesis of GO resulted in an increased interlayer spacing, from 3.37 Å in NFG to approximately 7.4 Å, indicating the introduction of various functional groups through oxidation. Subsequent thermal reduction of GO yielded reduced graphene oxide (rGO), which exhibited a slightly larger interlayer spacing along the c-axis compared to bulk graphite. SEM images of GO (Fig. 3a–h) revealed a two-dimensional sheet-like structure with multiple lamellar layers. These sheets had a thickness of 1–2  $\mu\text{m}$ , larger than that of single-layer graphene, primarily due to the presence of oxygen-containing functional groups. The hydrophilic nature of GO facilitated its uniform deposition onto substrates as thin films. SEM images of rGO, obtained through thermal reduction of GO, displayed ultra-thin graphene films with a thickness of less than  $\sim 10$  nm. Analysis using DSC/TGA indicated that GO experienced mass loss below 100 °C, attributed to the evaporation of adsorbed water and pyrolysis of oxygen-containing groups. The thermal stability of rGO exceeded that of GO, evident from its higher onset decomposition temperature and complete decomposition at a higher temperature range. FTIR spectroscopy analysis confirmed the presence of oxygen-containing functional groups in GO, which were significantly reduced in rGO following thermal treatment. The UV–vis spectra of aqueous dispersions of GO and rGO exhibited characteristic features corresponding to plasmon peaks and transitions of aromatic C–C bonds. Overall, this study successfully synthesized GO using the modified Hummers method and demonstrated the thermal reduction process to obtain rGO. The characterization and analysis of the materials provided valuable insights into their structural, thermal, and optical properties.



**Fig. 1** SEM micrograph of **a** Graphite, **b** GO12H, **c** GONP, **d** GOSIM [33]

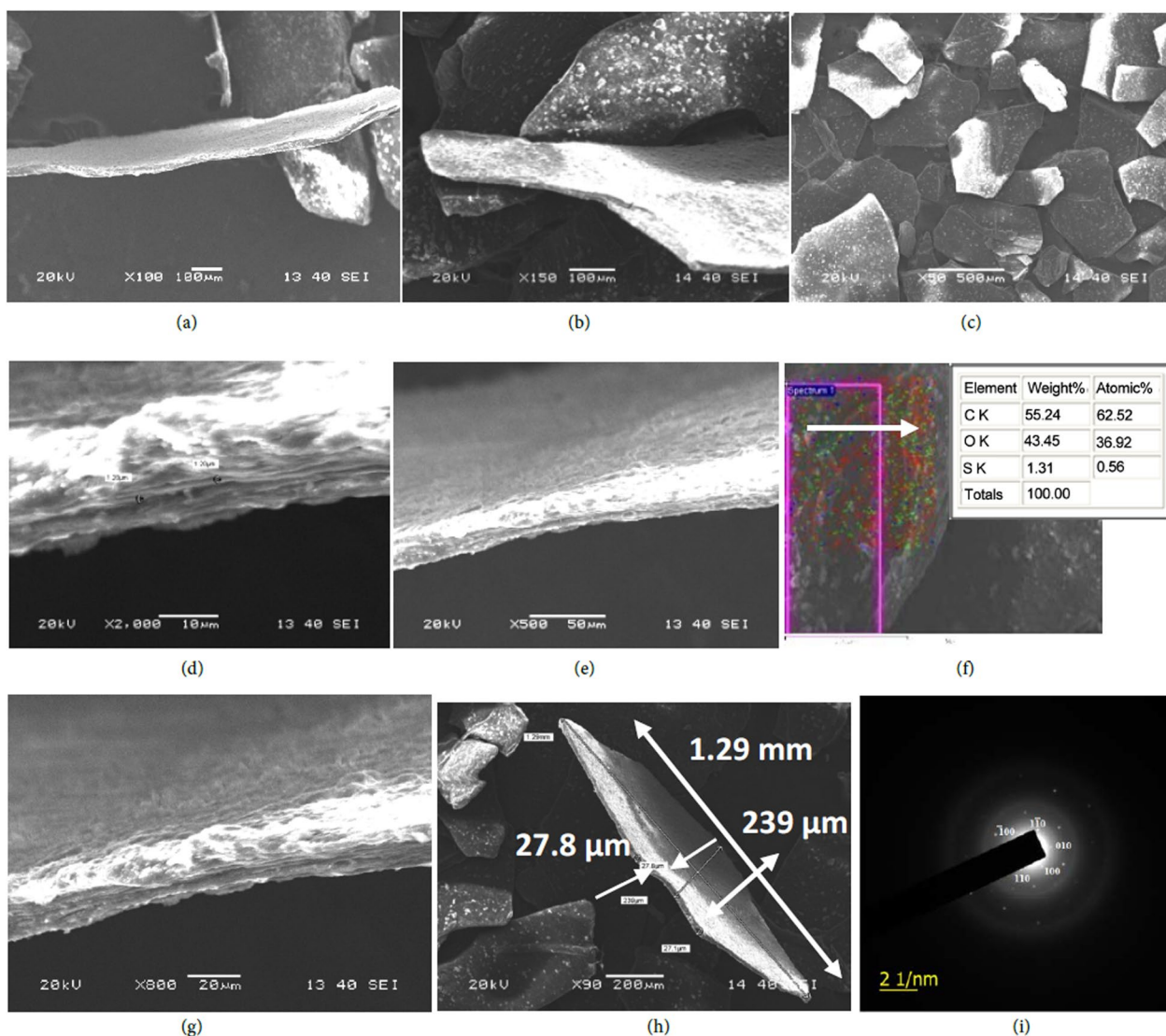


**Fig. 2** a–c SEM images of natural flake graphite (NFG) [34]

**Summary of the properties of graphene oxide synthesized using the Hummers method**

The typical characteristics attributed to graphene oxide synthesized via the Hummers method encompass:

- i. High oxygen content: GO synthesized using the Hummers method contains a high concentration of oxygen functional groups, such as hydroxyl, carboxyl, and epoxy groups [35, 36]. These functional



**Fig. 3** a–h SEM and EDX of GO [34]

groups make GO highly hydrophilic and can be used to modify its surface properties for various applications [36–39].

- ii. Thickness: GO synthesized using the Hummers method typically has a thickness of a few nanometers to a few micrometers. The thickness can be controlled by adjusting the synthesis conditions such as the concentration of oxidizing agents and reaction time [40–44].
- iii. Surface area: GO synthesized using the Hummers method has a high specific surface area, which makes it suitable for applications such as adsorption and catalysis [45–49].
- iv. Electrical conductivity: GO synthesized using the Hummers method has a lower electrical conduc-

tivity compared to pristine graphene due to the presence of oxygen functional groups, which act as electron-donating groups. However, the electrical conductivity of GO can be improved by reducing the oxygen content through thermal or chemical reduction methods [50–54].

- v. Optical properties: GO synthesized using the Hummers method has unique optical properties due to its strong light absorption in the ultraviolet and visible regions. This property has led to the development of GO-based photodetectors and solar cells [55–59].

In general, these research studies show that the Hummers method is efficient in synthesizing graphene oxide

(GO) with a substantial level of oxidation and a layered or sheet-like structure. By modifying the synthesis parameters, such as reaction time, temperature, and oxidizing agent concentration, it is possible to tailor the physical, chemical, and electronic characteristics of the resulting GO material.

#### Synthesis of graphene oxide using Staudenmaier method

The Staudenmaier method is similar to the Hummers method but uses a mixture of nitric acid, sulfuric acid, and potassium chlorate. The potassium chlorate acts as a catalyst for the oxidation reaction and results in higher yields of GO. However, this method can also produce toxic chlorine gas and is therefore not as commonly used as the Hummers method. The Staudenmaier method involves the use of strong oxidizing agents to convert graphite into GO.

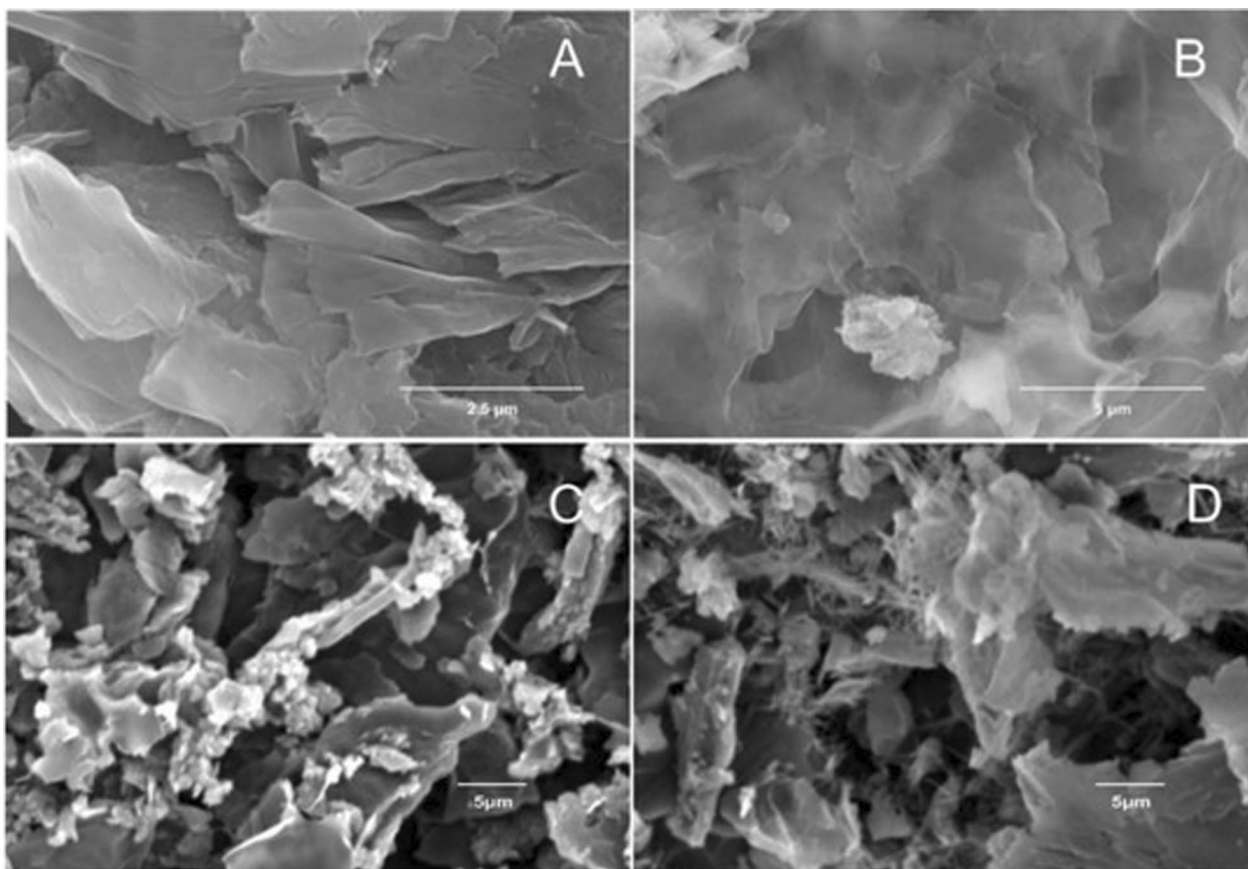
In the synthesis of graphene oxide (GO) using the Staudenmaier method [60–62], high-quality graphite flakes are employed as the initial material. Concentrated sulfuric acid ( $H_2SO_4$ ) and nitric acid ( $HNO_3$ ) act as oxidizing agents, while potassium chlorate ( $KClO_3$ ) assists in the oxidation process. The graphite flakes are combined with the acid solution in a reaction vessel, stirred at a controlled temperature, and heated for several hours. Gradual addition of  $KClO_3$  takes place during this period. Following oxidation, the reaction mixture is rapidly cooled by pouring it into either deionized water or an acidic solution. The resulting mixture is then filtered to gather the solid product, which is graphene oxide. Multiple washings with water or solvents are performed to eliminate impurities from the collected GO. The purified GO can be dried and subjected to characterization techniques such as SEM, TEM, XRD, FTIR, and Raman spectroscopy to evaluate its structural and chemical properties. It is crucial to observe appropriate safety precautions, including working in a well-ventilated environment, wearing protective gear, and complying with local regulations, due to the use of hazardous chemicals in the Staudenmaier method.

Lazanas et al. conducted a study where they reported the synthesis of hybrid materials incorporating reduced graphene oxide (rGO) and conjugated copolymers [63]. The researchers varied the number and arrangement of thiophenes in the copolymer's main chain (either indacenothiophene or indacenothienothiophene) as well as the type of polymer acceptor (difluoro benzothiadiazole or diketopyrrolopyrrole). They employed Stille aromatic coupling to synthesize the semiconducting copolymers and characterized them to determine their molecular properties. Graphene oxide was synthesized using the Staudenmaier method and subsequently transformed into reduced graphene oxide prior to conducting

structural analyses. They prepared different mixtures with varying amounts of rGO and conjugated copolymers to investigate the optoelectronic, thermal, and morphological characteristics.

SEM measurements were employed to examine the morphology of graphene oxide (GO), reduced graphene oxide (rGO), and the hybrid materials. Figure 4 illustrates the disparities between the nanosheets of GO (Fig. 4A) and rGO (Fig. 4B) in their dispersion forms, while Fig. 4C and D depicts the solid forms, revealing irregularities in rGO morphology attributed to the reduction process. The SEM images confirm the successful conversion of graphite to graphene oxide while maintaining a lamellar structure and demonstrate notable differences between film and solid forms due to the re-dispersion of GO and rGO in *o*-DCB. In Fig. 5, the SEM micrograph and elemental mapping analysis showcase a hybrid material composed of copolymer 2 (C2) with 5 wt% rGO. The interaction between the copolymer and rGO results in the intercalation of the copolymer within graphene nanosheets, as evidenced by the presence of carbon (Fig. 5B), sulfur (Fig. 5C) from the copolymer, and oxygen (Fig. 5D) from rGO. Comparable outcomes were observed for different copolymers containing 3, 5, and 10 wt% rGO, indicating the successful non-covalent attachment of rGO to the copolymers, even at lower concentrations. The results of UV–Vis spectroscopy revealed an increase in the maximum absorbance by approximately 3 to 6 nm for all hybrid materials, irrespective of the rGO concentration, compared to the pristine conjugated copolymers. This observation indicated distinct optical properties in the hybrid materials. The oxidation and reduction potentials were determined through voltammetric experiments, and the calculated HOMO and LUMO levels indicated a decrease in the electrochemical energy gap for low concentrations of rGO. Overall, the study highlights the potential of hybrid materials comprising graphene oxide and conjugated copolymers with high band gaps for applications related to organic solar cells.

In a 2012 study, Poh et al. examined various methods for preparing graphene, specifically the Staudenmaier, Hofmann, and Hummers methods, along with subsequent thermal exfoliation [27]. These techniques are commonly employed in the large-scale production of graphene sheets. The authors investigated the electrochemical characteristics of the graphene samples obtained through these methods. The Staudenmaier, Hofmann, and Hummers methods are chemical approaches that involve the oxidation and exfoliation of graphite. Each method has its own unique reaction conditions and reagents, resulting in variations in the properties of the resulting graphene materials. The researchers



**Fig. 4** SEM images of the GO and rGO in film (A, B) and in solid (C, D) forms

emphasized the significant differences observed in the electrochemical behavior of the graphene samples produced using these methods. Given graphene's potential applications in fields such as energy storage and sensing, its electrochemical properties are of great interest. The authors utilized techniques like cyclic voltammetry and chronoamperometry to analyze the electrochemical behavior of the graphene samples. They evaluated dissimilarities in electrochemical responses, including capacitive behavior, charge storage capacity, and redox activity, among the graphene materials prepared using the different methods. The study's findings demonstrated that the Staudenmaier, Hofmann, and Hummers methods, in combination with thermal exfoliation, yield graphene samples with distinct electrochemical properties. These differences were attributed by the authors to varying degrees of oxidation, defects, and functional groups introduced during the preparation processes. The results underscore the significance of selecting an appropriate synthesis method for graphene based on the desired electrochemical properties for specific applications.

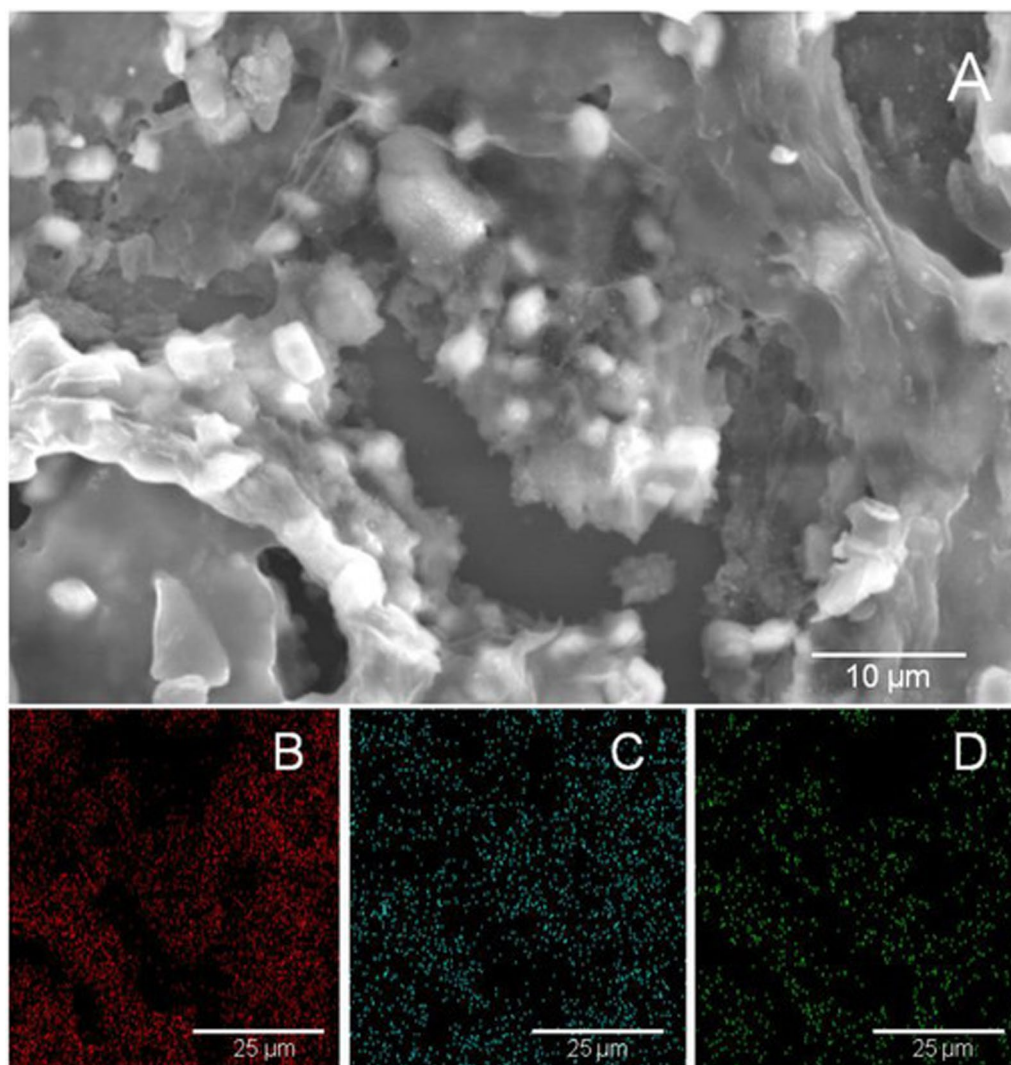
In summary, although both studies share the commonality of synthesizing graphene oxide using the

Staudenmaier method, their focus and specific outcomes vary. Lazanas et al. center their research on investigating hybrid materials derived from this synthesis method [63]. Conversely, Poh et al. undertake a comparative analysis of the electrochemical properties of graphene oxide, encompassing different synthesis methods, including Staudenmaier [27].

#### The properties of graphene oxide synthesized using the Staudenmaier approach

The properties typically associated with graphene oxide synthesized through the Staudenmaier method include:

- i. Increased production efficiency: Researchers have explored modifications to the Staudenmaier method to enhance the production efficiency of graphene oxide. This includes optimizing reaction conditions, such as temperature, reaction time, and the concentration of oxidizing agents, to achieve higher yields of graphene oxide [64–66].
- ii. Improved control over oxygen content: The Staudenmaier method allows researchers to control the oxygen content in graphene oxide by adjusting



**Fig. 5** SEM images of a hybrid system with the Copolymer 2 and the rGO (A). There is an apparent intercalation that is supported with the aid of EDS, where the elemental mapping analysis confirms the existence of carbon (B), sulfur (C) and oxygen (D)

- the reaction parameters. By optimizing the oxidation process, it is possible to obtain graphene oxide with desired oxygen functionalities, which influences its properties and potential applications [67–69].
- iii. Enhanced dispersion in solvents: One challenge with graphene oxide is its tendency to agglomerate or form restacked sheets, limiting its potential applications. Researchers have made progress in improving the dispersibility of Staudenmaier-synthesized graphene oxide in various solvents, allowing for better incorporation into different matrices and facilitating its use in composite materials [70–72].
  - iv. Tailored structural properties: The Staudenmaier method has been utilized to tailor the structural properties of graphene oxide. By controlling the oxidation parameters, such as the concentration of oxidants or the reaction time, researchers have achieved graphene oxide with different degrees of oxidation, layer thickness, and functional groups. These modifications influence the material's electrical, thermal, and mechanical properties [73–75].
  - v. Functionalization strategies: Staudenmaier-synthesized graphene oxide has been functionalized with various organic and inorganic compounds to introduce specific properties or enhance its compatibility with different matrices. Functionalization meth-



ods include covalent and non-covalent approaches, enabling the incorporation of graphene oxide into a broader range of applications, such as sensors, energy storage devices, and biomedical applications [76–79].

### Synthesis of graphene oxide using Tour method

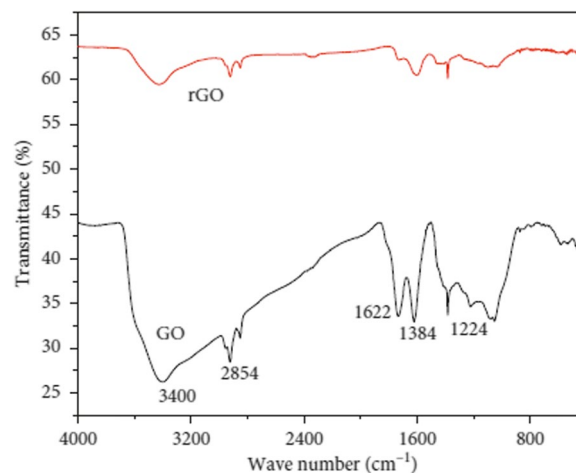
The Tour method, developed by Professor James Tour and his team at Rice University, is a specific approach for synthesizing graphene oxide (GO) [64, 65]. This method involves the chemical exfoliation of graphite to obtain GO sheets [13]. The process includes the oxidation of graphite using a strong oxidizing agent like a mixture of concentrated sulfuric acid ( $H_2SO_4$ ) and nitric acid ( $HNO_3$ ), followed by additional processing steps to achieve the desired graphene oxide material. The Tour method is known for its efficiency in producing high-quality GO with controlled properties suitable for various applications [13]. It employs a plasma torch to generate oxygen-containing functional groups on the graphite, resulting in GO with a higher degree of graphitization and a lower concentration of oxygen functional groups compared to GO synthesized using the Hummers method. However, it should be noted that the Tour method is more expensive and requires specialized equipment.

The modified Tour method was employed by Kotsyubynsky et al. to synthesize a colloidal solution of graphene oxide (GO), wherein the pH of the reaction medium was increased to 2.0–2.2 during the final stage of graphite oxidation by the addition of NaOH solution [80]. The resulting solid-phase graphene oxide consisted of multilayered graphene particles measuring approximately 7.5 nm in thickness (equivalent to 9–10 layers of graphene) and an average size of around 7.7 nm.

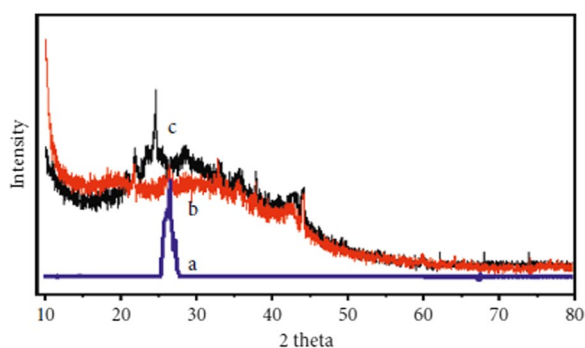
To obtain reduced graphene oxide (rGO), a combination of hydrazine and microwave reduction techniques was utilized. A comparative analysis of the structure, morphology, and electrical transport properties of rGO samples obtained through different methods was conducted, employing XRD, SAXS, Raman spectroscopy, low-temperature nitrogen adsorption, and impedance spectroscopy. The structural examination revealed two fractions of plate-like rGO particles for each reduction method, comprising stacks of 4–6 graphene layers and exhibiting lateral dimensions ranging from 7.1 to 7.6 nm. The microwave-reduced rGO exhibited a higher BET specific surface area ( $296\text{ m}^2\text{ g}^{-1}$ ) compared to the chemically reduced counterpart ( $237\text{ m}^2\text{ g}^{-1}$ ). The AC conductivity of both GO and rGO samples was investigated over a frequency range of  $10^{-2}$ – $10^5$  Hz at temperatures between 25 and 175 °C. The conductivity mechanism in the GO sample was predominantly

governed by proton exchange. For rGO samples obtained via chemical and microwave routes, a Drude-like response of electrical conductivity was observed at frequencies above  $10^3$  Hz, transitioning to Jonscher's law response at 175 °C. Changes in activation energies and relaxation times were interpreted using a model involving thermally activated, frequency-dependent electron hopping between randomly interconnected conducting  $sp^2$  rGO packages, separated by disordered  $sp^3$  rGO regions with correspondingly higher resistivity.

In another study, Habte and Ayele synthesized graphene oxide (GO) by immersing graphite in concentrated acid along with an oxidizing agent using the Tour's method [81]. Chemical reduction using ascorbic acid (AA) was employed to reduce graphene oxide, serving as both a reducing agent and a protecting agent. This process offered economic benefits and exhibited non-toxic and environmentally friendly characteristics. The oxidation level of graphite powder was assessed by varying parameters such as reaction time, temperature, and the quantity of  $KMnO_4$ . Graphene oxide (GO) and reduced graphene oxide (rGO) were characterized using various techniques, including visual observation, UV–Vis spectroscopy, FTIR spectroscopy, and X-ray diffraction (XRD). UV–Vis spectroscopy indicated the formation of GO and rGO, with maximum absorption peaks at 226 nm and 257 nm, respectively. FTIR spectroscopy spectra revealed the presence of oxygen-containing functional groups in GO after oxidation, which were subsequently removed during reduction (see Fig. 6). XRD analysis demonstrated that the diffraction peak of GO ( $2\theta = 10^\circ$ ) shifted to  $2\theta = 23.8^\circ$  (diffraction peak of rGO), indicating a change in the interlayer distance after reduction (see Fig. 7). This change was



**Fig. 6** FTIR spectra of GO and rGO [81]



**Fig. 7** X-ray diffraction patterns of a graphite powder, b GO powder, and c rGO powder [81]

attributed to the removal of oxygen-containing functional groups and the agglomeration of reduced graphene oxide sheets.

Based on the study's overall results, the most favorable oxidation level was achieved by treating 0.5 g of graphite with 4.5 g of  $\text{KMnO}_4$  (1:9 wt/wt ratio) in a mixture of 9:1  $\text{H}_2\text{SO}_4/\text{H}_3\text{PO}_4$  (v/v) for 12 h at a reaction temperature of  $50^\circ\text{C}$ .

#### Summary of the properties of graphene oxide synthesized using the Tour approach

The typical characteristics attributed to graphene oxide synthesized via Tour's method encompass:

- i. **Controlled oxidation:** The Tour method allows for precise control over the oxidation process, leading to uniform and well-defined graphene oxide sheets. This control enables researchers to tailor the properties of GO for specific applications [70, 80].
- ii. **Tunable functionalization:** The Tour method allows for the introduction of different functional groups on the GO surface. This functionalization can be customized, enabling the incorporation of specific properties such as enhanced solubility, improved dispersibility, or targeted chemical reactivity [82, 83].
- iii. **Size and thickness control:** The Tour method enables the production of graphene oxide with controlled size and thickness. This control is crucial for various applications where the dimensions of GO sheets play a significant role, such as in nanoelectronics, composite materials, and energy storage [84, 85].
- iv. **Enhanced stability:** Tour-synthesized graphene oxide has shown improved stability against aggregation and restacking compared to other synthesis methods. The controlled oxidation and functionalization processes help to minimize the tendency of

GO sheets to form irreversible aggregates, which is advantageous for the production of thin films and coatings [86, 87].

- v. **Tailored electrical and optical properties:** The introduction of oxygen-containing functional groups on the graphene surface modifies its electrical and optical properties. Tour-synthesized GO has demonstrated tunable electrical conductivity and optical absorption characteristics, making it suitable for applications in sensors, optoelectronics, and energy conversion devices [88, 89].

#### Synthesis of graphene oxide using electrochemical method

The synthesis of graphene oxide (GO) through an electrochemical method is based on the electrochemical exfoliation of graphite in the presence of an electrolyte [90]. This technique is known for its simplicity and ability to produce high-quality GO with a significant level of graphitization [91]. However, it does require specialized equipment and is not as widely utilized as other methods. Essentially, the electrochemical oxidation process converts graphite into graphene oxide by introducing oxygen-containing functional groups on its basal plane and edges, enabling its excellent solubility in water and polar solvents [92]. This approach offers a straightforward and scalable means of generating graphene oxide with carefully controlled properties. The electrochemical synthesis of graphene oxide has found extensive utility across diverse domains, such as energy storage, catalysis, sensors, and biomedical applications [93]. It acts as a fundamental building block for generating reduced graphene oxide (rGO) through subsequent reduction techniques. These processes reintroduce portions of graphene's  $\text{sp}^2$  carbon network, thereby augmenting its electrical conductivity [94].

The electrochemical synthesis of graphene oxide typically involves the following steps [90–94]:

- i. **Preparation of the graphite electrode:** A graphite electrode is prepared by polishing and cleaning a graphite substrate to ensure a clean and smooth surface.
- ii. **Electrolyte preparation:** An appropriate electrolyte solution is prepared, typically consisting of a mixture of a strong oxidizing agent and a supporting electrolyte. Common oxidizing agents include sulfuric acid ( $\text{H}_2\text{SO}_4$ ), nitric acid ( $\text{HNO}_3$ ), or a mixture of both (known as the Hummers method). The supporting electrolyte, such as sodium nitrate ( $\text{NaNO}_3$ ) or potassium permanganate ( $\text{KMnO}_4$ ),

helps in maintaining the electrochemical cell's conductivity.

- iii. Electrochemical cell assembly: The prepared graphite electrode is immersed in the electrolyte solution, and a counter electrode (usually a platinum or gold electrode) is placed in the same solution. The two electrodes are connected to an external power supply to establish an electrochemical cell.
- iv. Electrochemical oxidation: An electric potential is applied between the graphite electrode (anode) and the counter electrode (cathode). The graphite electrode undergoes electrochemical oxidation, resulting in the formation of graphene oxide. The electrochemical process involves the generation of reactive oxygen species (such as hydroxyl radicals) that react with the graphite surface, leading to the introduction of oxygen functional groups and the exfoliation of graphene layers.
- v. Collection and purification: After the desired electrochemical oxidation time, the graphene oxide is collected from the electrolyte solution. It can be isolated through various methods, including filtration, centrifugation, or sedimentation. The collected graphene oxide is then washed with water and other solvents to remove residual electrolyte and impurities.

The properties of the synthesized graphene oxide can be tailored by adjusting the electrochemical parameters such as applied potential, electrolyte composition, and oxidation time. These parameters influence the degree of oxidation, the concentration of functional groups, and the lateral size of the graphene oxide sheets.

In a recent study by Loudiki et al. an efficient direct electrochemical method was developed for the exfoliation of graphene from pencil graphite at ambient temperature [95]. This method was conducted using a small-scale lab apparatus, enabling practical implementation. The resulting graphene mixture, comprising graphene oxide (GO) and reduced graphene oxide (rGO), underwent comprehensive physical and electrochemical characterization. The application of an AC voltage of 11V (at 50 Hz) led to the initial formation of graphene oxide, followed by an increase in the reduction rate, which was observed to have a time-dependent effect. The researchers evaluated the performance of the prepared graphene through various assessments, including charge transfer, peak shape, and reversibility analysis of the  $[\text{Fe}(\text{CN})_6]^{3-/4-}$  redox system. The modified electrode exhibited remarkable electrochemical performance, owing to the notable enhancement in electron transport resulting from higher surface defects after electrode modification. The relationship between the reduction degree of graphene oxide and

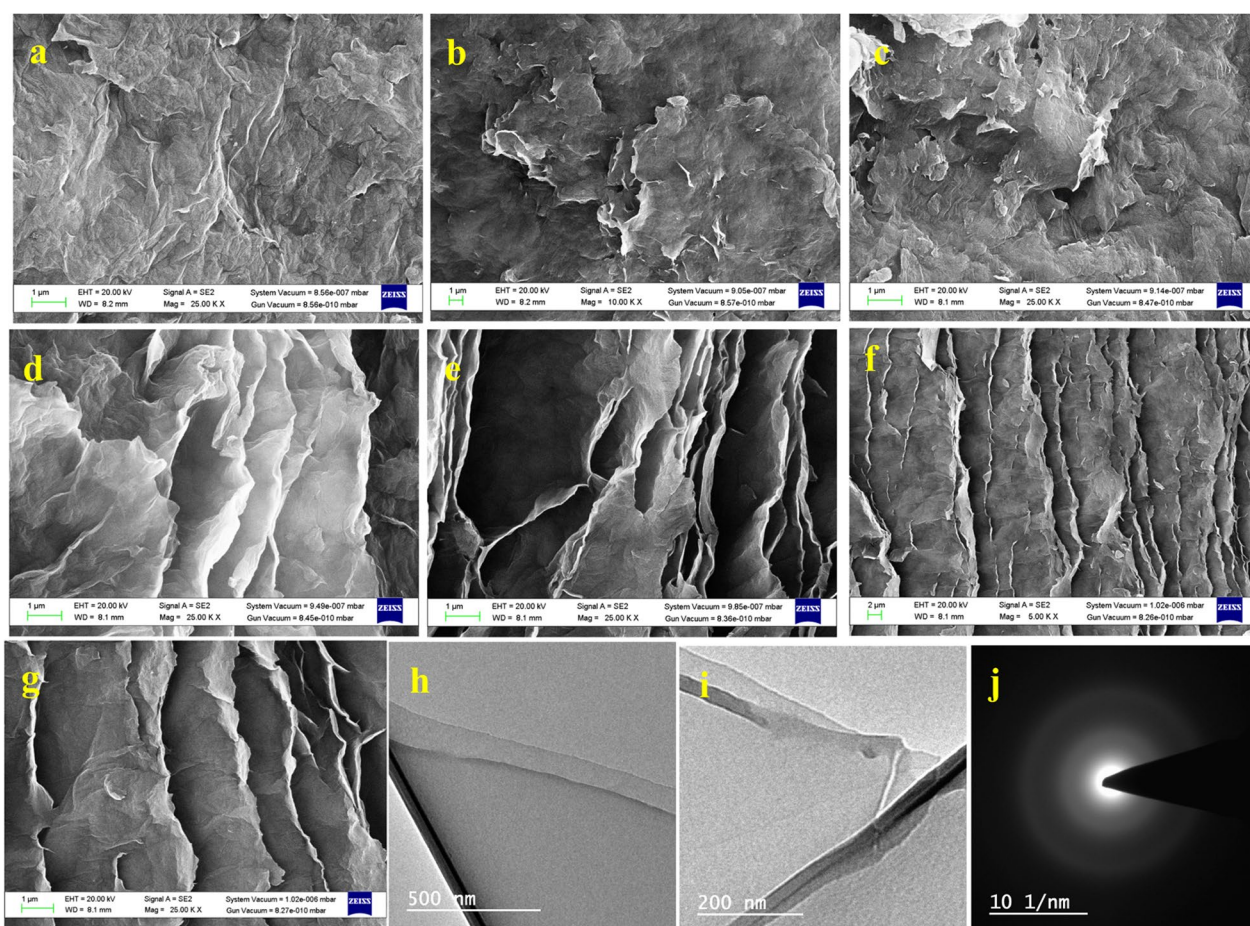
the interface and diffusion charge resistance exhibited an inverse correlation with the applied potential. This suggests that as the potential is adjusted, the interface and diffusion charge resistance change accordingly.

In another study, Kumar and Srivastava presented a straightforward two-step method to synthesize large graphene oxide (GO) sheets measuring approximately 10  $\mu\text{m}$  or larger [96]. The first step involved electrochemical exfoliation of a graphite electrode in a mixture of  $\text{H}_2\text{SO}_4$  and  $\text{H}_3\text{PO}_4$ , resulting in the production of graphene. In the second step, the exfoliated graphene sheets were oxidized using  $\text{KMnO}_4$  as the oxidizing agent. The oxidation process was carried out at around 60  $^\circ\text{C}$  for varying durations ranging from 1 to 12 h. The prepared GO samples underwent comprehensive characterization using various spectroscopy and microscopy techniques. X-ray diffraction (XRD), field emission scanning electron microscopy (FE-SEM), transmission electron microscopy (TEM), Fourier-transform infrared spectroscopy (FTIR), UV-visible spectroscopy, Raman spectroscopy, and thermogravimetric analysis were employed to assess the properties and quality of the synthesized GO. The UV-visible absorption spectrum of the GO samples exhibited a prominent peak at 230 nm and an adjacent band at 300 nm, corresponding to  $\pi-\pi^*$  and  $n-\pi^*$  transitions in all the samples. The relative percentages of oxygen-containing functional groups in the GO samples were determined using normalized FTIR plots, with GO synthesized for 6 h showing the highest content. Boehm titration was employed to quantify the functional groups on the surface of the GO. The XRD pattern of the GO synthesized for 6 h exhibited a characteristic peak at  $2\theta = 8.88^\circ$ , indicating an interplanar spacing of 0.995 nm between the layers. This result was considered one of the best compared to previous methods reported in the literature [97]. Raman spectroscopy revealed that the degree of defect (ID/IG) area ratio for the 6-h GO sample was 1.24, higher than that obtained using the widely used Marcano's approach. Field emission scanning electron microscopy (FE-SEM) analysis was performed to examine the morphologies of the different GO samples. The SEM images showed that GO sheets synthesized for 1 and 2 h had poorly exfoliated wrinkled structures, whereas GO samples synthesized for 6 to 12 h exhibited well-exfoliated sheet-like morphology with curled edges. Energy-dispersive X-ray (EDX) measurements determined the carbon-to-oxygen (C/O) ratios for the various GO samples, with GO synthesized for 6 h having a ratio of 0.94. Transmission electron microscopy (TEM) images of the 6-h GO sample confirmed the presence of thin sheets. The crystallinity of the GO sheets was assessed using selected area electron diffraction (SAED), which displayed diffused concentric diffraction rings, indicating

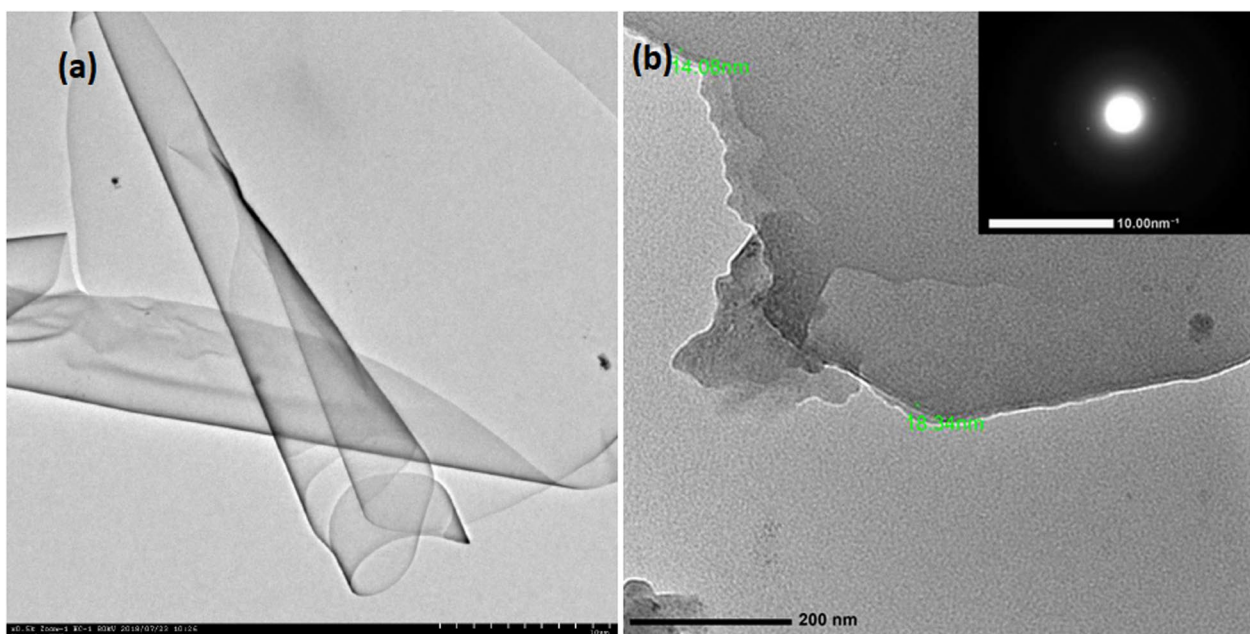
a low crystallinity of the material. Overall, the study demonstrated a simple and efficient approach to synthesize large GO sheets through electrochemical exfoliation and subsequent oxidation. The characterization techniques revealed the favorable properties and quality of the GO synthesized for 6 h, making it a promising material for various applications (see Fig. 8).

Singh et al. introduced a novel and environmentally friendly method for synthesizing reduced graphene oxide (rGO) [98]. Their approach involved electrochemical etching of carbon rods in a solution of lemon juice. The researchers conducted experiments using four different operating voltages to examine the impact on the quality of the synthesized rGO. Various analytical techniques, including Raman analysis and elemental analysis, were employed to investigate the differences among the rGO samples produced at different voltages. To assess the crystallite size variations and d-spacing,

the researchers analyzed the X-ray diffraction spectra using the Debye–Scherrer equation. The electron microscopy images revealed rGO sheets with folded edges, as depicted in Fig. 9. Additionally, a transmission electron microscopy (TEM) view of the rGO film demonstrated clear micrographs of stacked rGO layers with rolled edges (Fig. 9a). Another TEM image was captured while focusing on the edge of the stacked rGO sheets, illustrating the stacking of 3 to 4 rGO layers together (Fig. 9b). The thickness of these stacked sheets was determined to be approximately 14 nm. The formation of rGO and the increase in the number of layers with higher applied voltages were confirmed by UV–visible absorption spectra. Comparative studies indicated that the highest quality rGO was produced at a bias voltage of 10 V. The chemical-free and straightforward synthesis approach of this method suggests the potential for replacing other techniques for rGO production, with the possibility of further improvements.



**Fig. 8** FE-SEM images of GO corresponding to reaction times of **a** GO (1 h), **b** GO (2 h), **c** GO (4 h), **d** GO (6 h), **e** GO (8 h), **f** GO (10 h), and **g** GO (12 h); **h**, **i** TEM images of GO (6 h); and **j** SAED pattern of GO (6 h) [96]



**Fig. 9** TEM image of **a** rGO film in 10-micron range with folded edges, **b** stacked rGO sheets, (inset) SAED graph of rGO10 sample [98]

#### Summary of the properties of graphene oxide synthesized using electrochemical method

The properties typically associated with graphene oxide synthesized through the electrochemical method encompass:

- i. Scalability: electrochemical methods have shown promise in scaling up the production of graphene oxide. By using larger electrode setups and optimizing the electrochemical reaction parameters, researchers have been able to synthesize graphene oxide in larger quantities, facilitating its potential commercial applications [93, 94].
- ii. Improved purity and structural control: Electrochemical methods have been utilized to improve the purity and structural control of graphene oxide. By carefully controlling the reaction conditions, such as electrolyte composition, pH, temperature, and applied voltage, researchers have been able to enhance the homogeneity and consistency of the synthesized graphene oxide sheets [94, 95].
- iii. Reduced defects: Electrochemical methods have shown potential in reducing the number of defects in graphene oxide structures. Defects can negatively impact the properties of graphene oxide, such as its electrical conductivity and mechanical strength. Through the optimization of electrochemical parameters, researchers have been able to minimize the defect density, leading to graphene oxide with improved properties [95, 96].

- iv. Tailored functional groups: Electrochemical methods have allowed for the introduction of specific functional groups onto the graphene oxide surface. By modifying the electrolyte composition or employing electrochemical functionalization techniques, researchers have been able to incorporate desired functional groups onto the graphene oxide structure. This functionalization enhances the chemical reactivity and enables the targeted application of graphene oxide in various fields [96–98].

Table 1 presents a comparative analysis of the synthesis of graphene oxide using Hummers method, Tour method, and the electrochemical method, focusing on their individual advantages and disadvantages [27, 34–98]:

#### Diverse approaches for removing emerging contaminants

The approaches for removing emerging contaminants can vary significantly based on the specific contaminant and the chosen treatment method. Here are some common strategies for eliminating emerging contaminants:

- A. Adsorption: adsorption is widely employed to eliminate emerging contaminants from water or air. It involves the attachment of contaminants to a solid surface, like activated carbon or other adsorbent materials. Through physical or chemical interactions, contaminants adhere to the surface of the adsorbent,

**Table 1** Comparative analysis of graphene oxide synthesis methods

Method	Hummers method	Tour method	Electrochemical method
Principle	Chemical oxidation using strong oxidants and acids	Organic synthesis and covalent functionalization of graphene with reactive organic molecules	Electrochemical reduction of graphite oxide
Advantages	Widely used, well-established method	Allows precise functionalization of graphene sheets, easy to control the degree of functionalization	Simple experimental setup, mild reaction conditions
Disadvantages	Harsh reaction conditions (strong acids, high temperature)	Time-consuming and multi-step process	Requires specialized equipment (electrochemical cell, electrodes), limited scalability
Scalability	Moderate	Limited scalability due to the complex synthesis steps and functionalization processes	Limited scalability due to the need for specialized equipment and slower reaction kinetics
Degree of functionalization	Highly oxidized graphene oxide with numerous oxygen functional groups	Moderate degree of functionalization, controlled by the choice of reactive organic molecules	Depends on the applied potential and reaction time
Structural control	Limited control over the size and thickness of graphene oxide sheets	Can tune the functional groups and density of functionalization on graphene	Limited control over the thickness of reduced graphene oxide
Applications	Energy storage devices, composites, sensors, and coatings	Functionalized graphene for targeted applications like biosensors, drug delivery systems, and electronics	Energy storage devices, sensors, catalysts, and electrochemical electrodes

effectively extracting them from water or air [99–103].

- B. Catalytic degradation: catalytic degradation uses catalysts to facilitate the breakdown of emerging contaminants. These catalysts can be either heterogeneous (solid-phase catalysts) or homogeneous (liquid-phase catalysts). Contaminants react with the catalyst, decomposing into simpler, less harmful substances [104, 105].
- C. Membrane filtration: membrane filtration, a separation process, employs a semi-permeable membrane to separate contaminants from water or other liquids. The membrane contains small pores or channels allowing water molecules to pass while blocking larger contaminants. Various membranes, such as reverse osmosis (RO) or nanofiltration membranes, effectively remove a wide array of emerging contaminants, including micropollutants and nanoparticles [106, 107].
- D. Other relevant approaches: Additional strategies for removing emerging contaminants exist, contingent on specific contaminants and treatment methods:
- Advanced oxidation processes (AOPs): AOPs utilize highly reactive species, like hydroxyl radicals, to oxidize and degrade contaminants. Techniques such as ozonation, photocatalysis, or electrochemical oxidation achieve AOPs [108].
  - Biological treatment: Biological processes, including biodegradation or bioremediation, use microorganisms to break down contaminants into harmless byproducts via metabolic processes [109].
  - Chemical precipitation: Chemical precipitation involves adding chemicals to induce the formation of insoluble precipitates. These precipitates are then separated from water through sedimentation or filtration, commonly used for the removal of heavy metals or metalloids [110, 111].
  - Ion exchange: Ion exchange involves swapping ions between a solid-phase ion exchange resin and water. The resin selectively adsorbs certain ions, including emerging contaminants, releasing less harmful ions in exchange [112].
  - Physical separation: Certain emerging contaminants are removed through physical separation methods such as sedimentation, coagulation, or flocculation. These processes aggregate or settle the contaminants, facilitating their removal from water [113, 114].

It is crucial to note that the selection of the appropriate approach depends on specific contaminants, their concentration, and the desired treatment objectives. Often, a combination of multiple approaches is

employed in water or wastewater treatment processes to efficiently remove emerging contaminants.

### **Effectiveness of graphene oxide in removing emerging contaminants**

Graphene oxide (GO) has shown effectiveness in removing emerging contaminants from various environmental matrices [115]. Several studies have investigated the adsorption properties of GO towards different types of emerging contaminants, including pharmaceuticals, personal care products and pesticides. The unique physicochemical properties of GO contribute to its adsorption capabilities and make it a promising material for water treatment applications [116].

The large surface area and high adsorption capacity of GO enable it to effectively capture and remove emerging contaminants from water [117]. The  $\pi$ - $\pi$  stacking interactions, hydrogen bonding, and electrostatic attractions between GO and contaminants facilitate their adsorption onto the GO surface. Additionally, the presence of oxygen-containing functional groups on GO, such as hydroxyl and carboxyl groups, enhances its adsorption capacity through additional interactions with the contaminants [117].

The adsorption efficiency of GO can be influenced by various factors, including the properties of the contaminants (e.g., molecular size, polarity, and charge), the concentration of contaminants in the water, pH, temperature, and contact time. Optimization of these parameters can enhance the removal efficiency of GO for specific contaminants [115].

### **Graphene oxide (GO) as an efficient adsorbent for the removal of contaminants from water**

There have been several case studies that have explored the effectiveness of graphene oxide (GO) in removing emerging contaminants. One example is the removal of pharmaceutical compounds from water.

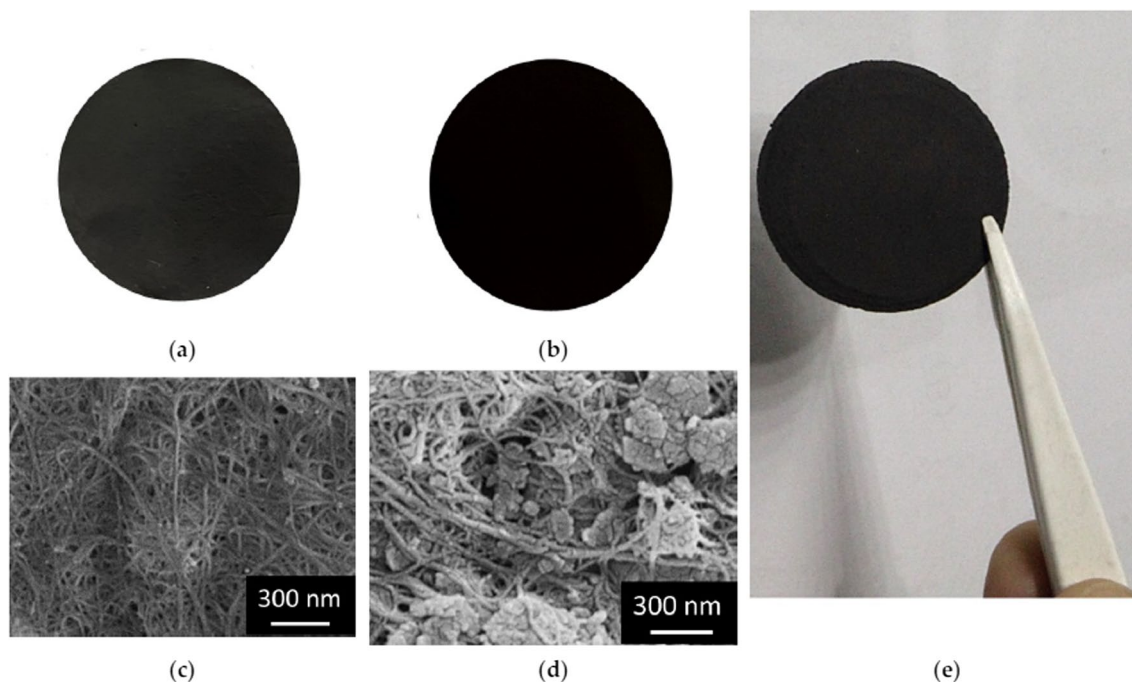
In a study carried out by Banerjee et al. the effectiveness of graphene oxide nanoplatelets (GONPs) in removing ibuprofen from water was investigated [115]. The researchers characterized the GONPs using electron microscopy and X-ray diffraction to analyze any changes in structure and morphology caused by the adsorption process. Batch adsorption experiments were conducted to assess the impact of various process parameters on the percentage removal of ibuprofen. The obtained data were analyzed using isotherm and kinetic analysis to understand the distribution of ibuprofen between the liquid and solid phases in the batch studies. The researchers found that the Langmuir isotherm model best described the adsorption behavior, and the process

followed pseudo-second-order kinetics. Thermodynamic parameters, including Gibbs' free energy, enthalpy, and entropy, were also evaluated. The results indicated that the adsorption of ibuprofen onto graphene oxide was an endothermic and spontaneous process. Based on their findings, the authors concluded that graphene oxide could serve as a suitable adsorbent for the efficient treatment of water contaminated with ibuprofen and similar anti-inflammatory drugs on a larger scale. The study highlights the potential of graphene oxide as an effective adsorbent for pharmaceutical removal from water.

Another case study was carried out by Baratta et al. focused on evaluating the efficacy of graphene oxide/single-walled carbon nanotube composite membranes (GO-SWCNT BPs) in removing three non-steroidal anti-inflammatory drugs (NSAIDs)—diclofenac, ketoprofen, and naproxen [116]. Various parameters were investigated, including pH conditions, graphene oxide content, and initial concentrations of NSAIDs. SEM analysis of the BP membranes revealed a consistent appearance with black and stable membranes, an average thickness of around  $100 \pm 2 \mu\text{m}$ , and an average diameter of  $38 \pm 1 \mu\text{m}$  (Fig. 10a, b). The SEM images also exhibited clusters and bundles of single-walled carbon nanotubes (SWCNTs) due to intermolecular interactions (Fig. 10c). The membranes demonstrated high permeability and large contact surface area, indicating effective adsorption.

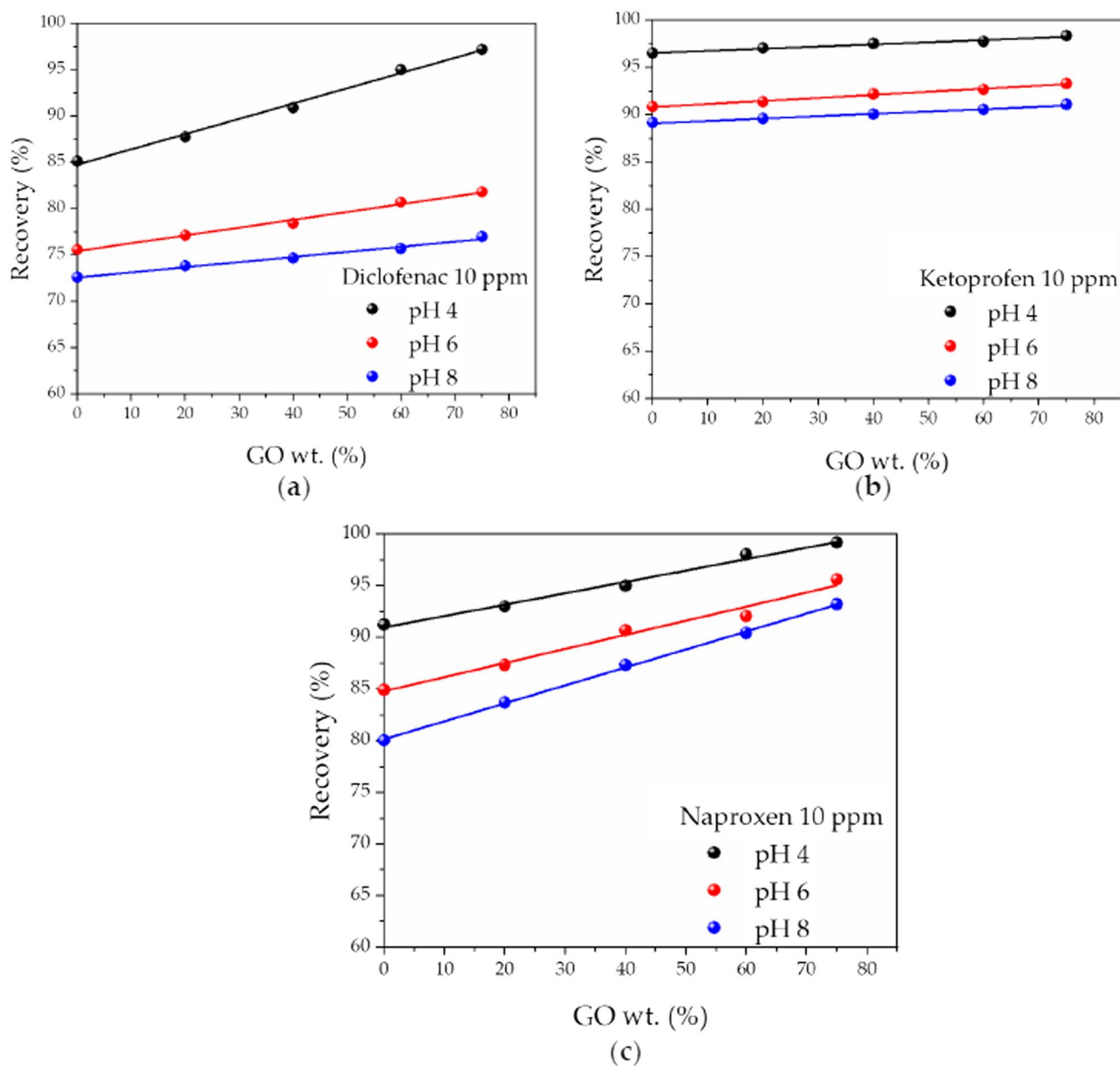
Incorporation of graphene oxide within the SWCNT BP membranes was observed, resulting in a homogeneous distribution of GO sheets (Fig. 10d). The adsorption capacities of the GO-SWCNT BPs were influenced by the graphene oxide content, with the highest capacities achieved at 75 wt.% graphene oxide, specifically  $118 \pm 2 \text{ mg g}^{-1}$  for diclofenac,  $116 \pm 2 \text{ mg g}^{-1}$  for ketoprofen, and  $126 \pm 3 \text{ mg g}^{-1}$  for naproxen at pH 4 (Fig. 11). Overall, the study suggests that GO-SWCNT BPs offer a promising and cost-effective approach for the removal of NSAIDs from drinking water sources. The membranes possess characteristics such as easy recovery and reusability, making them a viable solution for addressing the presence of NSAIDs in water resources.

In a previous study, Nodeh et al. developed a magnetic graphene oxide-based adsorbent, GO-MNPs-SiO<sub>2</sub>, to enhance the removal of naproxen from wastewater [117]. The incorporation of magnetic nanoparticles and silica onto graphene oxide improved water permeability, prevented sheet aggregation, and facilitated easy recovery using an external magnet. The adsorbent was synthesized, characterized, and applied for naproxen removal from sewage samples. The researchers investigated the effect of GO-MNPs-SiO<sub>2</sub> dosage on naproxen removal, varying the adsorbent mass from 5 to 160 mg. The results showed that increasing the dosage from 5 to 30 mg led to an increase in removal efficiency from 20 to 90%, with



**Fig. 10** Pictures of **a** SWCNT BP and **b** 75% GO-SWCNT BP. SEM images of **c** SWCNT BP and **d** 75% GO-SWCNT BP. The presence of GO flakes embedded in the SWCNT BP network is evident in the latter image. Samples up to 75 wt.% GO were self-sustainable and flexible disks **(e)** [116]

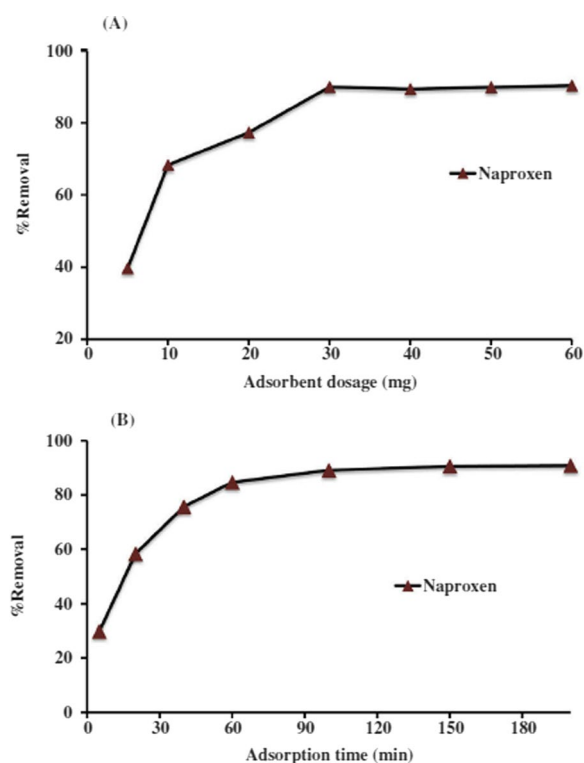




**Fig. 11** The recovery percentage,  $Re(\%)$ , of: **a** diclofenac, **b** ketoprofen, and **c** naproxen water solutions at different pH values and 10 ppm initial concentration as a function of GO content in GO-SWCNT BPs [116]

a slight stabilization at 30–60 mg (Fig. 12a). The influence of contact time on adsorption was also examined, and it was observed that the removal efficiency increased from 25 to 90% as the time increased from 5 to 60 min (Fig. 12b). This indicated a rapid adsorption process occurring before reaching equilibrium. To understand the adsorption mechanism, the experimental data were fitted to Langmuir, Freundlich, and D-R isotherms, and free energy calculations were performed. The maximum adsorption capacity of the nanocomposite was determined to be  $31.25 \text{ mg g}^{-1}$  at pH 5 and a contact time of

60 min. The Freundlich model provided the best fit to the data ( $R^2=0.999$ ), suggesting multilayer adsorption with a physisorption mechanism for naproxen removal. The calculated free energy value ( $0.49 \text{ kJ mol}^{-1}$ ) further supported this finding. In general, the GO-MNPs-SiO<sub>2</sub> nanocomposite exhibited high sorption capacity and fast kinetics for the removal of naproxen from aqueous solutions. The study concluded that it holds promise as an effective adsorbent for wastewater treatment applications in the removal of naproxen and similar pollutants.



**Fig. 12** Effect of amount of a adsorbent dosage and b contact (adsorption) time on the naproxen removal efficiency [117]

Additionally, there have been other investigations that have explored the utilization of graphene oxides in addressing the contamination of water by personal care products and pesticides. The accumulation of pesticide residues in the environment has become a significant concern due to their persistence and stability. As a result, researchers have shown renewed interest in finding effective methods to eliminate pesticides from the environment, employing a range of biological and chemical approaches [118]. One particular approach that has gained considerable attention is the use of graphene oxide (GO) for enhancing the adsorption of pesticides in aqueous solutions. GO has shown impressive adsorption capabilities, making it a promising candidate for environmental applications, especially in water purification to remove pesticide residuals [120]. However, despite numerous studies highlighting the adsorption potential of GO for environmental contaminants, our understanding of the underlying molecular mechanisms involved in this process is still limited.

Therefore, the primary objective of the case studies mentioned below is to evaluate the efficacy of graphene oxides in addressing the specific types of contaminants mentioned, namely pesticides. By conducting these case studies, researchers aim to delve deeper into the

adsorption mechanisms of graphene oxides and gain a better understanding of their effectiveness in tackling pesticide contamination. This knowledge is crucial for guiding future research and development efforts, as well as for providing insights into the potential applications of graphene oxide in environmental remediation and water purification processes.

The study conducted by Wang et al. focused on investigating the adsorption mechanism of pesticides by graphene oxide (GO) using fully atomistic molecular dynamics (MD) simulation and density functional theory (DFT) calculations [119]. The researchers aimed to gain a deeper understanding of the interactions between GO and pesticides, shedding light on the fundamental mechanisms involved in pesticide adsorption and providing insights for potential future applications. By employing advanced computational techniques, Wang et al. discovered that the major adsorption interactions between GO and pesticides were attributed to two key factors:  $\pi$ - $\pi$  stacking and van der Waals interactions.  $\pi$ - $\pi$  stacking refers to the stacking of aromatic rings, which is a common interaction observed in organic systems. Van der Waals interactions, on the other hand, are non-covalent forces that arise between molecules or molecular fragments due to fluctuations in electron density. The integration of density functional theory calculations, fully atomistic molecular dynamics simulations, and binding free energy calculations allowed the researchers to comprehensively explore and analyze the adsorption process. By doing so, they not only filled the theoretical gap in understanding the adsorption mechanisms of GO but also provided valuable insights into visualizing the adsorption process itself. These insights are crucial for guiding future applications of GO in adsorption processes. In general, the study by Wang et al. [119] made significant contributions by elucidating the underlying mechanisms of pesticide adsorption by GO. The findings not only enhanced our theoretical understanding of the adsorption process, but also paved the way for the development of improved adsorption applications involving GO.

The effect of graphene-based structures on the elimination of chlorpyrifos and dimethoate from water was investigated by Lazarević-Pašti et al. [120]. The study revealed that the adsorption of pesticides onto graphene-based adsorbents was highly dependent on the structural properties of both the sorbent and the sorbate. Interestingly, the surface area of the adsorbent was found to be less influential in controlling the removal efficiency.

The researchers observed that the aliphatic pesticide dimethoate exhibited a preference for adsorption onto hydrophilic oxidized graphene surfaces. On the other hand, the graphene basal plane, which possesses a  $\pi$

electron system and high structural order, selectively removed chlorpyrifos due to its aromatic moiety. Moreover, the presence of an intermediate concentration of oxygen functional groups on the surface of the graphene-based adsorbent led to effective elimination of both chlorpyrifos and dimethoate.

More recently, the use of graphene oxide (GO) as an adsorbent has been investigated for the removal of various toxic pesticides, including atrazine, ametryn, chlorpyrifos, and malathion, from water and wastewater [121–123]. GO nanoplatelets have been applied for the adsorption of chloridazon and its degradation metabolites (desphenyl-chloridazon: DC and methyl-desphenyl-chloridazon: MDC) from aqueous solutions [124]. The study established the good adsorption capacity of GO for chloridazon ( $67.18 \text{ mg g}^{-1}$ ), DC ( $34.30 \text{ mg g}^{-1}$ ), and MDC ( $36.85 \text{ mg g}^{-1}$ ), which was attributed to efficient remediation through hydrogen bonding, hydrophobic interactions, and  $\pi$ - $\pi$  interactions.

In addition to experimental adsorption studies, theoretical investigations have explored the performance of graphene-based materials for the adsorption of pesticides, such as atrazine [125]. Graphene and its defected forms, including hexagonal boron nitride and carbon-doped hexagonal boron nitride, have been studied using density functional theory (DFT) calculations. The molecular orbital interactions and energy calculations provided insights into the affinity of graphene, hexagonal boron nitride, and carbon-doped hexagonal boron nitride towards atrazine and estimated the magnitude of the interaction.

The results indicated that physical interactions, particularly dispersive interactions, played a significant role in stabilizing the adsorbed atrazine. The mobility of adsorbed atrazine on graphene adsorbents was affected by relatively low rotational and translational energy barriers. In contrast, in hexagonal boron nitride, the movement of atrazine was limited by electrostatic attractions and polar bonds due to the high translational energy barrier.

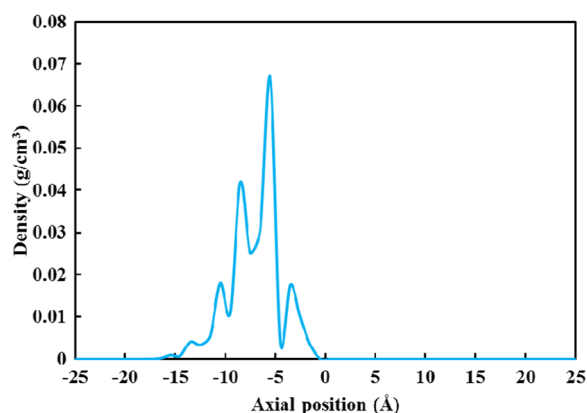
These findings highlight the potential of graphene-based materials, including graphene oxide and its derivatives, in the removal and remediation of various pesticides from water. Both experimental and theoretical investigations have provided valuable insights into the adsorption mechanisms and the potential applications of these materials for pesticide removal, contributing to the development of efficient and sustainable water treatment approaches.

The challenge of removing perchlorate from water, owing to its non-volatile, highly soluble, and kinetically inert nature, has led to the exploration of various treatment technologies. Among these methods, microbial

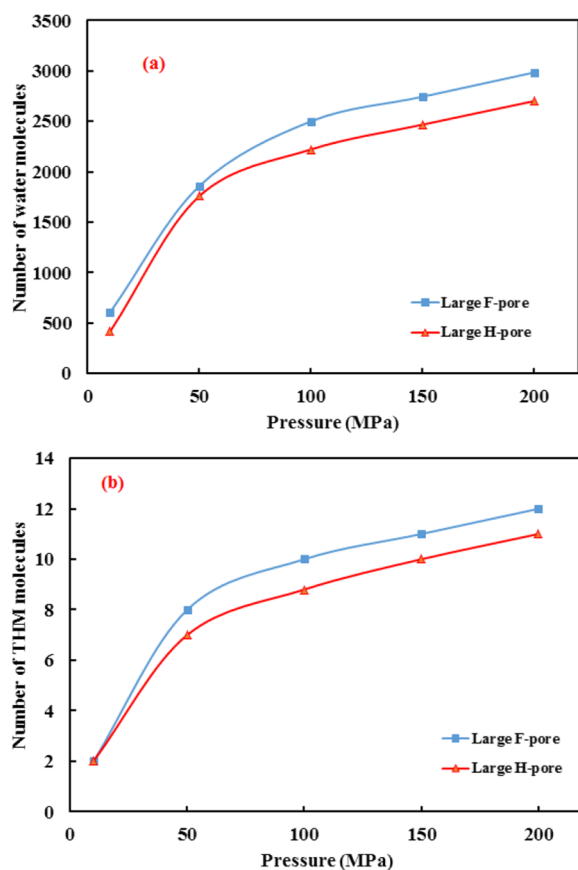
reduction, ion exchange, membrane technologies, chemical reduction, and adsorption have been investigated. Notably, adsorption, especially using carbon-based materials, has gained significant attention due to its effectiveness and cost efficiency in purifying water.

In a pivotal study conducted by Lakshmi and Vasudevan in 2013, a batch adsorption process was employed to investigate the removal of perchlorate ions ( $\text{ClO}_4^-$ ) from water using graphene [126]. The graphene, prepared through liquid-phase exfoliation, exhibited remarkable adsorption efficiency of 99.2%. Through extensive characterization techniques such as Raman spectroscopy, Fourier-transform infrared spectroscopy, and scanning electron microscopy, the researchers confirmed graphene's efficacy as an adsorbent for perchlorate removal. The study systematically explored the adsorption process by varying parameters such as pH, ionic strength, and temperature. The results indicated that graphene displayed a high perchlorate adsorption capacity of up to  $0.024 \text{ mg/g}$  at an initial concentration of  $2 \text{ mg/L}$  and a temperature of  $298 \text{ K}$ . Kinetic studies revealed that the second-order kinetics model best described the adsorption process, while the Langmuir adsorption isotherm accurately represented the equilibrium data. Crucially, thermodynamic studies demonstrated that the adsorption of perchlorate by graphene was a spontaneous and endothermic process. This innovative approach effectively reduced perchlorate concentrations in water to permissible levels, rendering it drinkable. The research showcased the potential of graphene-based materials in addressing the challenge of perchlorate contamination, paving the way for sustainable and efficient water purification solutions.

The presence of trihalomethanes (THMs) in drinking water, resulting from the reaction of natural organic substances with chlorine during water treatment, poses significant health risks. THMs, including  $\text{CH}_3\text{Cl}$ ,  $\text{CH}_2\text{Cl}_2$ , and  $\text{CHCl}_3$ , are known carcinogens and have been linked to kidney, liver, and nervous system problems. Therefore, effective separation methods are crucial. In a study conducted by Azamat et al. molecular dynamics simulations were utilized to investigate the permeability of Trihalomethanes (THMs) through graphene membranes under induced pressure conditions [127]. The researchers explored various graphene pore sizes and chemical terminations to understand the separation process in detail. Results from the simulations indicated that functionalized nanoporous graphene with smaller diameters effectively prevented the permeation of THMs while allowing water molecules to pass through. Specifically, in simulations with pore sizes below a certain threshold (as depicted in Fig. 13), only water molecules were observed to traverse the graphene, effectively blocking the passage



**Fig. 13** Density profile of all types of THMs in the small H-pore at a pressure of 200 MPa in z direction of system. (THMs remained on the one hand of graphene and could not pass across the small pores.)



**Fig. 14** **a** The number of water molecules passing through the large pores; **b** the number of THMs passing through the large pores. (Waters and THMs pass from the large pores.)

of THMs. Conversely, larger pores, as shown in Fig. 14, enabled the passage of both THMs and water molecules. Under specific pressure conditions, the simulations demonstrated a reduction in the concentration of THMs in the filtered water. For instance, at a pressure of 200 MPa, the concentration of THMs was reduced from the initial 0.4 to 0.037 mol/L. These findings highlight the potential of thin graphene membranes with tailored nanopores in efficiently removing THMs from water. The study's outcomes, provide crucial insights into the design and optimization of graphene-based filtration systems for addressing water contamination challenges caused by disinfection by-products.

Perfluorooctanoic acid (PFOA) and other per- and poly-fluoroalkyl substances (PFASs) have emerged as critical environmental pollutants due to their persistent nature and adverse health effects. Traditional methods for PFAS remediation are energy-intensive and often ineffective, necessitating the exploration of innovative in situ strategies. In a recent study by Lath et al. the focus shifted towards advanced adsorbents, specifically graphene oxide (GO), an iron oxide-modified reduced-GO composite (FeG), and RemBind™ (RemB), an activated-carbon/clay/alumina-based adsorbent [128]. The objective was to assess their efficacy in PFAS sorption and shed light on the underlying mechanisms, aiming to contribute to the development of efficient remediation technologies. Graphene oxide, known for its highly negative surface charge due to oxygen functional groups, initially seemed promising. However, its anionic nature posed a challenge, leading to repulsion with anionic PFAS species like PFOA. This limitation prompted the exploration of innovative modifications. FeG, a hybrid material resulting from iron-mineral-functionalization of GO, exhibited remarkable sorption capabilities. Its consistent high sorption (over 90%) across a range of pH values and ionic strengths indicated the dominance of non-electrostatic forces, suggesting the involvement of hydrophobic interactions and ligand-exchange mechanisms with associated Fe-minerals. The study also compared RemB, which showed similar efficiency to FeG, emphasizing the potential of combining mineral and carbon phases in remediation strategies.

Crucially, the research delved into desorption studies, providing valuable insights into the stability of adsorbed PFAS. Methanol, among the tested solvents, demonstrated the most effective desorption, indicating strong and relatively irreversible binding. This finding is pivotal for assessing the risk of PFAS remobilization, especially concerning environmental factors like rainfall events. Beyond laboratory conditions, FeG and RemB showcased their real-world applicability by effectively sorbing various PFASs from a contaminated-site water sample.

This success underscores the potential of these ‘mixed’ adsorbents in practical scenarios, opening avenues for enhanced sorption through multifaceted mechanisms. In summary, the study by Lath et al. [128] highlights the transformative potential of advanced adsorbents like FeG and RemB in addressing PFAS contamination. Their ability to offer high sorption capacities, stability against desorption, and versatility in real-world applications marks a significant step forward in the quest for sustainable, in situ PFAS-remediation technologies. As research in this field advances, the integration of diverse materials and innovative modifications promises a future where PFAS-contaminated sites can be effectively restored, ensuring environmental safety and public health.

**Comparing graphene oxide (GO) with traditional treatment methods for the removal of emerging contaminants: advantages and insights**

When comparing graphene oxide (GO) with traditional treatment methods for the removal of emerging contaminants, it becomes evident that GO exhibits several advantages. Traditional methods, such as activated carbon adsorption, advanced oxidation processes, and membrane filtration, have been widely used in water treatment, but they may have limitations in terms of efficiency, cost, or selectivity [126].

One of the key advantages of GO is its large surface area, which provides a significant adsorption capacity [127]. The abundant surface area allows GO to interact with and adsorb a wide range of contaminants, including emerging contaminants such as pharmaceuticals and pesticides [127, 128]. Additionally, GO can be easily functionalized, meaning that its surface properties can be modified to further enhance its adsorption capabilities. Functionalization can be achieved by introducing specific

functional groups or by doping the GO structure with other elements, tailoring it to target specific contaminants or improve selectivity [129].

Another advantage of GO is its ease of synthesis. GO-based adsorbents can be synthesized using relatively simple methods, making the production process scalable and cost-effective [130]. Moreover, GO-based materials have demonstrated good stability, which is crucial for their long-term performance in water treatment applications. Additionally, GO adsorbents have shown the potential for reusability, reducing waste generation and overall treatment costs [130].

In contrast, traditional treatment methods may face challenges in terms of their efficiency, cost-effectiveness, or selectivity [131, 132]. Activated carbon adsorption, for example, is widely used for water treatment but may have limitations in terms of its adsorption capacity and selectivity for specific contaminants [133]. Advanced oxidation processes, such as ozonation or photocatalysis, can be effective in degrading contaminants but may require additional energy consumption and produce potentially harmful byproducts [134, 135]. Membrane filtration, while efficient for the removal of particulate matter, may have limited effectiveness in removing dissolved organic contaminants [136, 137].

Given the advantages of GO, including its large surface area, high adsorption capacity, ease of functionalization, synthesis scalability, stability, and potential reusability, it holds significant promise as an alternative adsorbent material for removing emerging contaminants [138, 139]. Further research and development efforts are needed to optimize its performance, explore its potential synergies with other treatment methods, and address any potential challenges or limitations. Table 2 presents the advantages of graphene oxide (GO) over traditional treatment

**Table 2** The advantages of graphene oxide (GO) over traditional treatment methods for emerging contaminants

Parameters for comparison	Graphene oxide (GO)	Traditional treatment methods
Effectiveness in contaminant removal	High efficiency due to large surface area and high adsorption capacity	Variable effectiveness depending on the specific method and contaminant
Selectivity towards target contaminants	Can be tailored for specific contaminants through functionalization	Limited selectivity, often removing a wide range of contaminants
Adsorption capacity	High adsorption capacity, particularly for organic compounds	Varies depending on the material used; may require additional adsorbents for efficient removal
Degradation efficiency	Can exhibit catalytic properties for degradation of certain contaminants	Dependent on the specific treatment method; may require additional steps for degradation
Regeneration potential	Can be regenerated and reused for multiple cycles	Regeneration potential may vary depending on the treatment method
Cost-effectiveness	Production costs are decreasing; potential for cost-effective large-scale applications	Costs can vary depending on the treatment method and scale of implementation
Environmental impact	Potential for lower environmental impact; can be synthesized from sustainable sources	Environmental impact can vary depending on the specific treatment method and chemicals used

methods for emerging contaminants [10, 14, 126–136, 139–142].

#### Factors affecting removal efficiency of graphene oxide (GO) for emerging contaminants

Several factors can significantly influence the removal efficiency of graphene oxide (GO) for emerging contaminants. Understanding these factors is crucial for optimizing the performance of GO-based adsorbents and ensuring their effective application in water treatment processes.

- i. Contaminant characteristics: The physicochemical properties of the contaminants themselves play a crucial role in their adsorption onto GO [10, 14, 140–144]. Factors such as molecular size, polarity, solubility, and chemical structure can impact the interactions between the contaminants and GO surfaces [145]. Contaminants with larger molecular sizes or higher hydrophobicity tend to have stronger interactions with GO and exhibit higher adsorption capacities [146]. Additionally, the presence of functional groups in the contaminant molecules can facilitate specific interactions, such as hydrogen bonding or  $\pi$ - $\pi$  stacking, leading to enhanced adsorption onto GO [147]. For example, in the study conducted by Wu et al. the impact of contaminant characteristics, specifically the molecular size and the number of benzene rings, on graphene (G) and chemically reduced graphene oxide's adsorption performance was explored [148]. The researchers investigated the adsorption of various organic chemicals, including acrylonitrile (AN), p-toluenesulfonic acid (p-TA), 1-naphthalenesulfonic acid (1-NA), and methyl blue (MB), using graphene-based materials as adsorbents. The results revealed that organic chemicals with larger molecular sizes and more benzene rings exhibited higher adsorption speeds and maximum adsorption capacities on Graphene. Specifically, p-TA, 1-NA, and MB demonstrated maximum adsorption capacities of  $\sim 1.43$  g/g G,  $\sim 1.46$  g/g G, and  $\sim 1.52$  g/g G at 30 °C, respectively. These values were noted to be the highest among various nanomaterials studied for these contaminants. Additionally, the adsorption process for MB on graphene was found to be temperature-dependent, indicating that higher temperatures facilitated the adsorption. Furthermore, the study observed that graphene's efficiency remained stable during the initial five cycles of the adsorption–desorption process, indicating its potential for reusability. Fluorescence spectra analysis suggested

that the adsorption of MB on Graphene involved a  $\pi$ - $\pi$  stacking adsorption process. In a nutshell, the research demonstrated that graphene could serve as a promising adsorbent for the removal of chemicals containing benzene rings in wastewater. This finding underscores the significance of considering contaminant characteristics, such as molecular size and structure, when designing efficient adsorption processes using graphene-based materials.

- ii. GO properties: The properties of GO, including its surface area, surface chemistry, and functional groups, are critical for its adsorption performance. GO typically possesses a large surface area, which provides more binding sites for contaminants [149]. The presence of oxygen-containing functional groups, such as hydroxyl and carboxyl groups, on the GO surface contributes to its hydrophilicity and enhances the adsorption of polar contaminants [150, 151]. Additionally, the surface charge of GO can affect the electrostatic interactions with charged contaminants [150, 151]. For instance, Esteban-Arranz et al. demonstrated the significance of graphene oxides (GO) in removing contaminants from water [152]. They explored different GO types and observed key interactions using infrared spectroscopy. The study highlighted the vital role of GO properties, including surface area and chemistry, in determining adsorption performance. Cooperative effects between pollutants were noted with GO, emphasizing interlayer spaces' importance. Reduced graphene oxides showed synergetic interactions, with aromatic structures influencing adsorption capacity. Overall, the research underscores the complex relationship between GO properties and its effectiveness in contaminant removal.
- iii. Solution conditions: The adsorption behavior of contaminants onto graphene oxide (GO) is influenced by various conditions within the aqueous solution, including parameters such as pH, temperature, and ionic strength [153]. Specifically, the pH level of the solution plays a crucial role as it affects both the surface charge of GO and the ionization state of the contaminants, thereby influencing electrostatic interactions between them [154]. Additionally, temperature variations can impact the adsorption process kinetics and the stability of the GO adsorbent [155]. Changes in ionic strength can affect the competition between ions in the solution and the adsorption sites on GO, potentially altering the adsorption capacity. For example, the study conducted by Song et al. exemplifies how the adsorption behavior of contaminants

onto graphene oxide (GO) is intricately influenced by the conditions of the aqueous solution [156]. In their research, they activated ATP using high temperature and HCl, resulting in a-ATP, and subsequently prepared a GO/a-ATP composite through hydrothermal synthesis. Under specific conditions, including a dosage of 0.75 g/L, pH of 5, reaction time of 120 min, initial temperature of 35 °C, and an initial tetracycline (TC) concentration of 50 mg/L, the adsorption capacity of GO/a-ATP for TC reached 38.8 mg/g. The kinetic data analysis, using pseudo-first-order (PFO), pseudo-second-order (PSO), and mixed-order (MO) models, demonstrated that the adsorption process is chemisorption-driven, involving two rate-limiting steps: diffusion and adsorption, with diffusion being the predominant factor. Equilibrium data fitting revealed that the Langmuir model provided the best fit, indicating a homogeneous and monolayer adsorption process. Additionally, thermodynamic parameters like standard Gibbs free energy ( $\Delta G^\circ$ ) and standard enthalpy ( $\Delta H^\circ$ ) indicated an endothermic nature of the adsorption reaction. This study showcased the importance of optimizing solution conditions to enhance the adsorption efficiency of GO-based composites, providing valuable insights into the practical application of these materials in water purification processes.

- iv. Contact time: The contact time between the GO adsorbent and the contaminated water is an important factor affecting the adsorption process [157]. Longer contact times allow for more extensive interaction between contaminants and the GO surface, leading to increased removal efficiency [158]. For example, in the study by Falahati et al. the effect of contact time on the adsorption capacity of graphene oxide (GO) nanosheets for dairy wastewater treatment was explored [16]. The researchers varied the duration of contact between the adsorbent and wastewater to understand its influence on pollutant removal. They found that as the contact time increased, the removal efficiencies for pollutants such as total nitrogen (TN), total phosphorus (TP), chemical oxygen demand (COD), and turbidity also increased. The results indicated that a longer contact time allowed for a more thorough interaction between the GO nanosheets and the pollutants in the wastewater. As a result, the adsorption process had more time to occur, leading to higher removal efficiencies. This finding highlights the importance of sufficient contact time in maximizing the adsorption capacity of GO nanosheets, ensuring effective removal of contaminants from

dairy wastewater. Proper optimization of contact time is crucial in practical applications of GO-based adsorbents for efficient water treatment processes.

- v. Interference from coexisting substances: Within real-world water matrices, the presence of concurrent substances, which could include natural organic matter, inorganic ions, or competing contaminants, has the potential to disrupt the adsorption process of target contaminants onto graphene oxide (GO) [159]. These substances may compete for adsorption sites, block active sites on the GO surface, or reduce the effective surface area available for adsorption [8]. Understanding and mitigating these interferences are essential for achieving optimal removal efficiency. For example, by considering these factors and their interactions, researchers and engineers can optimize the design and operation of GO-based adsorption processes for the removal of emerging contaminants [160]. This knowledge can aid in the development of effective water treatment strategies that harness the unique properties of GO and address the challenges associated with emerging contaminants in water sources. For example, the study carried out by Yao et al. looked into the intricate dynamics of adsorbing typical antibiotics (tetracycline and sulfadiazine) and heavy metals [Cu(II) and Zn(II)] onto graphene oxides (GO), a promising nano-adsorbent [161]. The investigation included a comprehensive analysis of coexisting substances, such as natural organic matter, inorganic ions, and competing contaminants, that could potentially interfere with the adsorption process. The study revealed that the presence of heavy metals significantly enhanced the adsorption capacities of antibiotics onto GO, indicating a synergistic effect. This enhancement was attributed to the formation of complexes between heavy metals and GO, with heavy metals acting as “bridges” facilitating the adsorption process. In contrast, antibiotics had a minor promotional effect on the adsorption of heavy metals. Additionally, the study highlighted the inhibitory role of coexisting salt ions, especially  $\text{Ca}^{2+}$ , in the adsorption process. These ions, by interacting with GO, reduced the adsorption capacity. Furthermore, the presence of humic acid provided additional sites for heavy metal uptake but competed with antibiotics for adsorption sites, demonstrating the complex interplay of different coexisting substances during the adsorption process. Interestingly, even after six adsorption-release cycles, the re-adsorption capacities remained high,

suggesting the feasibility and durability of GO in the removal of such combined contaminants. This study sheds light on the challenges and opportunities in designing efficient adsorption processes for complex wastewater systems involving multiple contaminants and coexisting substances.

#### Limitations and challenges of graphene oxide in removing emerging contaminants from waste waters

Graphene oxide (GO) has gained significant attention as a potential material for removing emerging contaminants from wastewater due to its unique physicochemical properties. However, like any other technology, GO also has certain limitations and challenges that need to be considered [162–179].

- i. **Fouling and aggregation:** Fouling and aggregation of graphene oxide (GO) during water filtration poses substantial challenges, causing membrane fouling and reducing filtration efficiency [162]. To overcome this obstacle, researchers are investigating strategies such as surface modification and composite material synthesis [163, 164]. These approaches aim to enhance the stability and dispersion of GO, offering potential solutions for improving water treatment processes [162–164]. Addressing GO fouling and aggregation is pivotal for advancing effective water treatment strategies. In a study by Alam et al. researchers explored innovative approaches using two-dimensional nanomaterials like GO, reduced graphene oxide (rGO), and molybdenum disulfide ( $\text{MoS}_2$ ) to modify conductive polymer surfaces [165]. By applying an external voltage, they investigated the attachment and release behavior of foulants on modified surfaces. The study revealed promising results: applying a negative voltage significantly delayed the attachment of foulants, demonstrating electrostatic repulsion. Moreover, the attachment rate reduced significantly on rGO-PPy and  $\text{MoS}_2$ -PPy surfaces compared to bare PPy under specific conditions. In the release study, NaCl electrolysis effectively removed deposited foulants from all surfaces, facilitated by the continuous generation of free chlorine during voltage application. This research offers valuable insights into mitigating fouling challenges in water filtration, paving the way for enhanced filtration technologies.
- ii. **Cost considerations:** The large-scale production of high-quality GO can be expensive, limiting its practical application in wastewater treatment. Graphene-based materials, including GO, often require complex and energy-intensive production methods, making them costly compared to traditional adsorbents. The cost-effectiveness of GO-based systems needs to be carefully evaluated to determine their feasibility for widespread implementation [166–170].
- iii. **Regeneration and disposal:** After adsorbing contaminants, the regeneration and disposal of GO-based adsorbents pose challenges [8]. Regeneration methods for GO typically involve desorption of contaminants using chemicals or heat treatment [171, 172]. However, these processes may not be efficient or sustainable, and they can lead to the generation of secondary pollutants or require additional energy consumption. The proper disposal of used GO-based adsorbents is also a concern, as the long-term environmental impacts of graphene-based materials are still being studied [171, 172]. For example, in the study by Sharif et al. two reduced graphene oxide/iron oxide (rGO-IO) nanocomposites were developed for efficient water treatment [171]. These nanocomposites, composed of magnetite as the iron oxide phase, allowed easy separation using a magnetic field. The nanocomposites exhibited promising adsorption properties, with a peak capacity of  $39 \text{ mg g}^{-1}$  for the composite with 60 wt% iron oxide, dropping to  $26 \text{ mg g}^{-1}$  at 75 wt% iron oxide. Electrochemical regeneration was explored due to rGO's high conductivity and nonporous surface. This method, energy-efficient and rapid, achieved 100% regeneration efficiency within 30 min at a current density of  $10 \text{ mA cm}^{-2}$ , enhancing capacity to  $80 \text{ mg g}^{-1}$  after the second cycle. However, it's crucial to address challenges linked to GO regeneration, often involving chemical or heat-based desorption methods. These conventional approaches pose sustainability issues, such as secondary pollutant generation and additional energy consumption. While the electrochemical method offers an efficient solution, long-term viability and environmental impact require further investigation. Sustainable disposal and prevention of secondary pollutant generation are key concerns, making the sustainability of regeneration processes a vital area for future graphene-based adsorption research [171, 172].
- iv. **Interactions with other substances:** GO's interactions with other substances present in wastewater can affect its adsorption performance. Natural organic matter (NOM) and inorganic ions, such as sulfates and chlorides, can compete with contaminants for adsorption sites on GO [173]. These interactions may lead to reduced adsorp-



tion efficiency or selectivity for specific emerging contaminants. Understanding and controlling the influence of various water matrix components on GO's adsorption behavior is crucial for optimizing its performance. For example, in the study conducted by Song et al. the complexity of GO's interactions with various components in wastewater was clear [174]. GO's efficiency in adsorbing polycyclic aromatic hydrocarbons (PAHs) and their oxygen derivatives (OPAHs) was remarkable; however, these interactions were influenced by the diverse substances present in the water solution. Natural organic matter (NOM) and inorganic ions, such as sulfates and chlorides, coexisted with the targeted contaminants. These substances competed with PAHs and OPAHs for adsorption sites on GO. The study highlighted the multifaceted nature of GO's interactions in a real-world wastewater environment. Despite the competitive presence of NOM and inorganic ions, the synthesized nitrogen-doped reduced graphene oxide (NRGO) exhibited exceptional adsorption efficiency for PAHs and OPAHs. The research not only showcased NRGO's potential but also underscored the importance of understanding and navigating GO's interactions with various wastewater constituents. This nuanced understanding is essential for optimizing GO-based adsorption processes in the complex and diverse setting of wastewater treatment, ensuring effective removal of emerging contaminants despite the challenges posed by coexisting substances.

- v. Scale-up and practical implementation: While the laboratory-scale studies have demonstrated the potential of GO in removing emerging contaminants, scaling up the technology for real-world applications remains a challenge [175–179]. Factors such as the development of cost-effective and efficient manufacturing processes, integration with existing wastewater treatment systems, and ensuring long-term stability and performance need to be addressed for practical implementation [175–179].

In summary, graphene oxide shows promise as an adsorbent for removing emerging contaminants from wastewater. However, its limitations and challenges, such as limited adsorption capacity, fouling and aggregation, cost considerations, regeneration and disposal, interactions with other substances, and scale-up issues, need to be thoroughly investigated and overcome to enable its successful application in large-scale wastewater treatment operations. Further research and development efforts are necessary to address these challenges

and fully exploit the potential of graphene oxide-based systems.

#### Toxicity of graphene oxide

The toxicity of graphene oxide (GO) is an important aspect to consider when exploring its potential applications in various fields, including biomedicine and environmental remediation. While GO offers numerous beneficial properties, its potential adverse effects on human health and the environment have raised concerns [180–190].

- i. Cellular toxicity: Studies have shown that GO can exhibit cytotoxicity towards various types of cells, including human lung cells, skin cells, and immune cells. The toxicity of GO is attributed to multiple factors, including its size, shape, surface chemistry, and concentration. The sharp edges and large surface area of GO sheets can cause physical damage to cell membranes, leading to cellular stress and inflammation. Additionally, the oxidative properties of GO, resulting from the presence of oxygen-containing functional groups on its surface, can induce the generation of reactive oxygen species (ROS), leading to oxidative stress and cell damage.
- ii. Inflammation and immune response: GO have been found to induce an inflammatory response in biological systems. When exposed to GO, immune cells release pro-inflammatory cytokines, leading to an inflammatory cascade. Prolonged or excessive inflammation can disrupt normal cellular functions and contribute to tissue damage. Moreover, GO can also interact with immune cells, such as macrophages, altering their morphology and function. These immune responses highlight the potential immunotoxicity of GO.
- iii. Genotoxicity: Some studies have reported genotoxic effects of GO, indicating its potential to cause DNA damage and induce mutations. The oxidative stress generated by GO can lead to DNA strand breaks, chromosomal aberrations, and DNA mutations. These genotoxic effects raise concerns about the potential long-term health risks associated with GO exposure.
- iv. Pulmonary effects: Inhalation exposure is a significant concern for GO due to its potential respiratory effects. Animal studies have shown that inhalation of GO can induce lung inflammation, fibrosis, and granuloma formation. The small size and high aspect ratio of GO sheets enable them to penetrate deep into the respiratory system, reaching the alveoli. Once in the lungs, GO can trigger inflammatory responses and cause tissue damage,

potentially leading to chronic respiratory conditions.

- v. Environmental impact: The environmental toxicity of GO is an emerging area of research. While GO is considered chemically stable, it can interact with organisms in the environment. Aquatic organisms, such as fish and algae, can be exposed to GO through wastewater discharge. Studies have shown that GO can affect the growth, development, and behavior of aquatic organisms. Additionally, the accumulation of GO in the environment may have long-term ecological consequences that need further investigation.

It is important to note that the toxicity of GO can be influenced by various factors, including its physicochemical properties, dispersion methods, concentration, exposure duration, and route of exposure. Researchers are actively working to understand the mechanisms underlying GO toxicity and develop strategies to mitigate its adverse effects. Surface modifications, functionalization, and the use of protective coatings are being explored to enhance the biocompatibility and reduce the toxicity of GO.

In summary, the toxicity of graphene oxide is a complex and multifaceted issue. While GO holds immense potential for various applications, including biomedicine and environmental remediation, its potential adverse effects on human health and the environment must be thoroughly assessed and addressed. It is crucial to conduct comprehensive toxicity studies and implement proper safety measures to ensure the safe and responsible use of graphene oxide-based materials.

#### **The environmental impact associated with the utilization of graphene oxide**

The environmental impact of graphene oxide (GO) use is an important consideration when assessing its potential applications in various fields. While GO offers unique properties and potential benefits, its potential adverse effects on ecosystems and the environment have raised concerns.

- i. Ecotoxicity to aquatic organisms: GO can enter aquatic ecosystems through wastewater discharge or accidental releases. Studies have shown that GO can have toxic effects on various aquatic organisms, including fish, crustaceans, and algae. The high surface area and reactive properties of GO can lead to physical and chemical interactions with organisms, affecting their behavior, growth, reproduction, and survival. Additionally, the potential accumulation and persistence of GO in aquatic environments may have long-term ecological consequences that require further investigation [191–194].
- ii. Soil and terrestrial ecosystem impacts: The potential release of GO into soil can affect soil quality and terrestrial ecosystems. Studies have shown that GO can influence soil microbial communities, affecting their composition and function. Soil organisms, such as earthworms, may be exposed to GO, potentially leading to adverse effects on their behavior and reproductive success. The long-term impacts of GO on soil ecosystems and the broader consequences for plant growth, nutrient cycling, and soil health require further research [194–197].
- iii. Effects on beneficial microorganisms: GO's antimicrobial properties, while potentially useful in certain applications, can also have unintended consequences. The broad-spectrum antimicrobial activity of GO may affect beneficial microorganisms involved in nutrient cycling, soil health, and plant–microbe interactions. Disruption of these microbial communities can have cascading effects on ecosystem functioning and stability [198–201].
- iv. Fate and transport in the environment: Understanding the fate and transport of GO in the environment is crucial to assess its potential environmental impact. Research has shown that GO can adsorb other pollutants, such as heavy metals and organic contaminants, which may affect their mobility and bioavailability in the environment. The aggregation and sedimentation of GO can result in its accumulation in sediments, potentially impacting benthic organisms and sediment-dwelling microbial communities [202, 203].
- v. Potential for bioaccumulation and biomagnification: The potential for GO to bioaccumulate and biomagnify in food chains is a concern. While studies have shown limited bioaccumulation of GO in certain organisms, the accumulation potential may vary depending on the species, exposure duration, and environmental conditions. If GO accumulates in higher trophic levels, it may pose risks to predators and organisms at the top of the food chain [204, 205].
- vi. Life cycle considerations: Assessing the environmental impact of GO requires considering its entire life cycle, including production, use, and disposal [22, 206]. The production of GO typically involves energy-intensive processes and the use of chemicals, which can contribute to greenhouse gas emissions and other environmental impacts [207, 208]. The disposal of GO-based products and waste should also be carefully managed to prevent

their release into the environment and potential long-term impacts [22, 206, 207].

### Future directions and challenges

The use of graphene oxide (GO) in the remediation of emerging contaminants holds great promise for addressing the growing challenges of environmental pollution. However, as with any emerging technology, there are future directions and challenges that need to be considered and addressed. In this discussion, we will elaborate exhaustively on the potential future directions and challenges of using graphene oxide in the remediation of emerging contaminants [176–196].

- i. Enhanced adsorption and degradation: Future research should focus on further enhancing the adsorption and degradation capabilities of graphene oxide for a broader range of emerging contaminants. This includes optimizing the properties of graphene oxide, such as surface chemistry, pore size, and functionalization, to improve its effectiveness in capturing and breaking down contaminants.
- ii. Selectivity and specificity: Developing graphene oxide-based materials with improved selectivity and specificity towards target contaminants is a crucial future direction. Tailoring the surface properties and functional groups of graphene oxide can enhance its ability to selectively adsorb or degrade specific pollutants while minimizing interference from other coexisting substances in the environment.
- iii. Scalability and cost-effectiveness: The scalability and cost-effectiveness of graphene oxide-based technologies are important considerations for real-world applications. Future efforts should focus on developing scalable synthesis methods for graphene oxide and exploring cost-effective production processes to make it commercially viable for large-scale remediation projects.
- iv. Long-term stability and reusability: Ensuring the long-term stability and reusability of graphene oxide-based materials is essential for sustainable remediation. Future research should aim to improve the stability of graphene oxide in different environmental conditions and develop effective regeneration techniques to restore its adsorption and degradation capacity for repeated use.
- v. Environmental fate and ecotoxicology: Understanding the environmental fate, transport, and potential ecotoxicological impacts of graphene oxide is a critical area of future research. Studying its behavior in different environmental compartments, such as water, soil, and sediments, as well as its effects on

non-target organisms, will help assess the overall environmental sustainability and safety of graphene oxide-based remediation strategies.

- vi. Integration with other technologies: Exploring the integration of graphene oxide with other emerging technologies, such as advanced oxidation processes, photocatalysis, or membrane filtration, can lead to synergistic effects and improved efficiency in the remediation of emerging contaminants. Future research should focus on developing innovative hybrid systems that combine the strengths of different technologies for enhanced remediation performance.
- vii. Field-scale applications: Demonstrating the effectiveness of graphene oxide-based remediation technologies at the field scale is a crucial step towards practical implementation. Future research should involve pilot-scale or full-scale studies to validate the performance, reliability, and feasibility of these technologies in real-world contaminated sites under varying environmental conditions.
- viii. Regulatory and policy considerations: The adoption of graphene oxide-based remediation technologies will require the development of appropriate regulations and policies. Future efforts should involve collaboration between researchers, regulators, and policymakers to establish guidelines for the safe and responsible use of graphene oxide, addressing potential concerns related to its environmental impact, health risks, and waste management.
- ix. Public perception and acceptance: Public perception and acceptance of graphene oxide-based remediation technologies play a significant role in their successful implementation. Future research should include public engagement activities, knowledge dissemination, and awareness campaigns to promote understanding, acceptance, and trust among stakeholders and the general public.
- x. Data sharing and standardization: Encouraging data sharing, collaboration, and standardization of methodologies and protocols within the scientific community will facilitate the advancement of graphene oxide-based remediation research. This will enable more comprehensive comparisons, reliable assessment of results, and the establishment of benchmarks for future studies.

In general, the use of graphene oxide in the remediation of emerging contaminants offers exciting opportunities, but it also faces several future directions and challenges. By addressing these challenges through ongoing research, technological advancements, and collaboration among various stakeholders, graphene oxide-based remediation

technologies can contribute significantly to addressing the complex environmental challenges associated with emerging contaminants.

## Conclusion

The remediation of emerging contaminants is of utmost importance due to their potential adverse effects on ecosystems and human health. Graphene oxide (GO) has emerged as a promising material for water treatment and the removal of emerging contaminants. This comprehensive review has provided an in-depth understanding of the synthesis methods, properties, mechanisms of contaminant removal, effectiveness, toxicity, environmental impact, and mitigation strategies associated with the use of graphene oxide. Graphene oxide exhibits unique structural and surface properties, including high surface area and porosity, chemical and mechanical stability, and catalytic activity, making it suitable for the removal of various emerging contaminants. The adsorption, catalytic degradation, and membrane filtration mechanisms employed by graphene oxide have demonstrated remarkable efficiency in removing contaminants such as pharmaceuticals and pesticides. Several case studies have highlighted the effectiveness of graphene oxide in removing emerging contaminants, often surpassing traditional treatment methods. Factors such as pH, contact time, concentration, and the presence of coexisting substances can influence the removal efficiency of graphene oxide. However, there are limitations and challenges associated with its use, including scalability, cost, regeneration, and the need for further research to optimize its performance. Toxicity studies have shown that graphene oxide can exhibit varying degrees of toxicity depending on its size, concentration, and surface functionalization. Furthermore, the environmental impact of graphene oxide use must be carefully managed through mitigation strategies such as sustainable production practices, waste management, and risk assessments. Future directions in the field of graphene oxide for the remediation of emerging contaminants involve exploring its potential for large-scale applications, integrating it with other treatment methods to enhance efficiency, addressing cost and feasibility challenges, and identifying knowledge gaps that require further research. Collaboration between researchers, regulators, and policymakers is crucial to developing appropriate regulations, policies, and public acceptance.

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## Author contributions

The primary manuscript text was authored by BA and IH I. MM, IEU and AIA were responsible for generating figures. The manuscript underwent review by all authors.

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The data and materials used in this study are available upon request.

## Declarations

### Ethics approval and consent to participate

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### Consent for publication

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