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Review

Advanced Graphene-Based Technologies for Antibiotic Removal from Wastewater: A Review (2016–2024)

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Abstract: The increasing presence of antibiotics in wastewater poses significant environmental risks, including the promotion of antibiotic resistance and harm to aquatic ecosystems. This study reviews advancements in graphene-based technologies for removing antibiotics from wastewater between 2016 and 2024. Graphene-based platforms, such as graphene oxide (GO), reduced graphene oxide (rGO), and graphene composites, have shown great promise in this field because of their exceptional adsorption capacities and rapid photocatalytic degradation capabilities. Functionalized graphene materials and graphene integrated with other substances, such as metal oxides and polymers, have enhanced performance in terms of antibiotic removal through mechanisms such as adsorption and photocatalysis. These technologies have been evaluated under various conditions, such as pH and temperature, demonstrating their practical applicability. Despite challenges related to scalability, cost-effectiveness, and environmental impact, the advancements in graphene-based technologies during this period highlight their significant potential for effective antibiotic removal, paving the way for safer and more sustainable environmental management practices.

Keywords: graphene; antibiotics; wastewater; photocatalytic degradation; adsorption

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1. Introduction

The increasing contamination of water sources with pharmaceutical pollutants, particularly antibiotics, poses significant environmental and public health challenges [1]. Antibiotics in wastewater contribute to the development of antibiotic-resistant bacteria [2], disrupt aquatic ecosystems [3], and enter the food chain [4], making their removal an urgent global priority. Conventional wastewater treatment methods [5], while effective at removing many contaminants, often fail to eliminate antibiotics, necessitating the development of more advanced treatment technologies.

Graphene-based materials (GBMs) have emerged as promising solutions for the removal of a wide range of pollutants because of their exceptional physicochemical properties, including high surface area, chemical stability, and unique adsorption capabilities [6]. Since its discovery, graphene and its derivatives, such as GO and rGO, have been extensively studied for environmental applications, particularly in water treatment [7]. The use of graphene-based technologies for antibiotic removal from wastewater has gained considerable attention because of their efficiency in adsorbing various pharmaceutical compounds, including antibiotics, and their potential for large-scale implementation [8].

This review aims to provide a comprehensive overview of the advancements in graphene-based technologies for the removal of antibiotics from wastewater from 2016–2024. By critically analyzing recent literature following the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) guidelines [9], this study highlights the various mechanisms involved in antibiotic adsorption, the role of graphene derivatives in enhancing removal efficiency, and the challenges associated with the practical implementation of these technologies. This review also identifies future research directions, emphasizing the need for more sustainable and economically viable solutions in wastewater treatment.

1.1. Graphene-Based Materials

Graphene, a two-dimensional (2D) form of carbon in which atoms are arranged in a hexagonal lattice, has emerged as one of the most promising materials in nanotechnology and materials science. As discovered in 2004 [10], graphene's unique combination of properties, such as exceptional electrical and thermal conductivity, mechanical strength, flexibility, and optical transparency, has led to its exploration in a wide range of applications, from electronics to energy storage and beyond [11]. These exceptional properties have spurred the development of various GBMs, each tailored to exploit specific aspects of graphene's capabilities. The diversity of these materials stems from modifications in the structure, composition, and synthesis processes of graphene, allowing for the customization of its properties to meet the demands of different technological applications [12].

1.1.1. Types of Graphene-Based Materials

GBMs can be classified into several categories based on their structure (Figure 1). Each type of graphene-based material offers unique properties that make it suitable for specific applications.

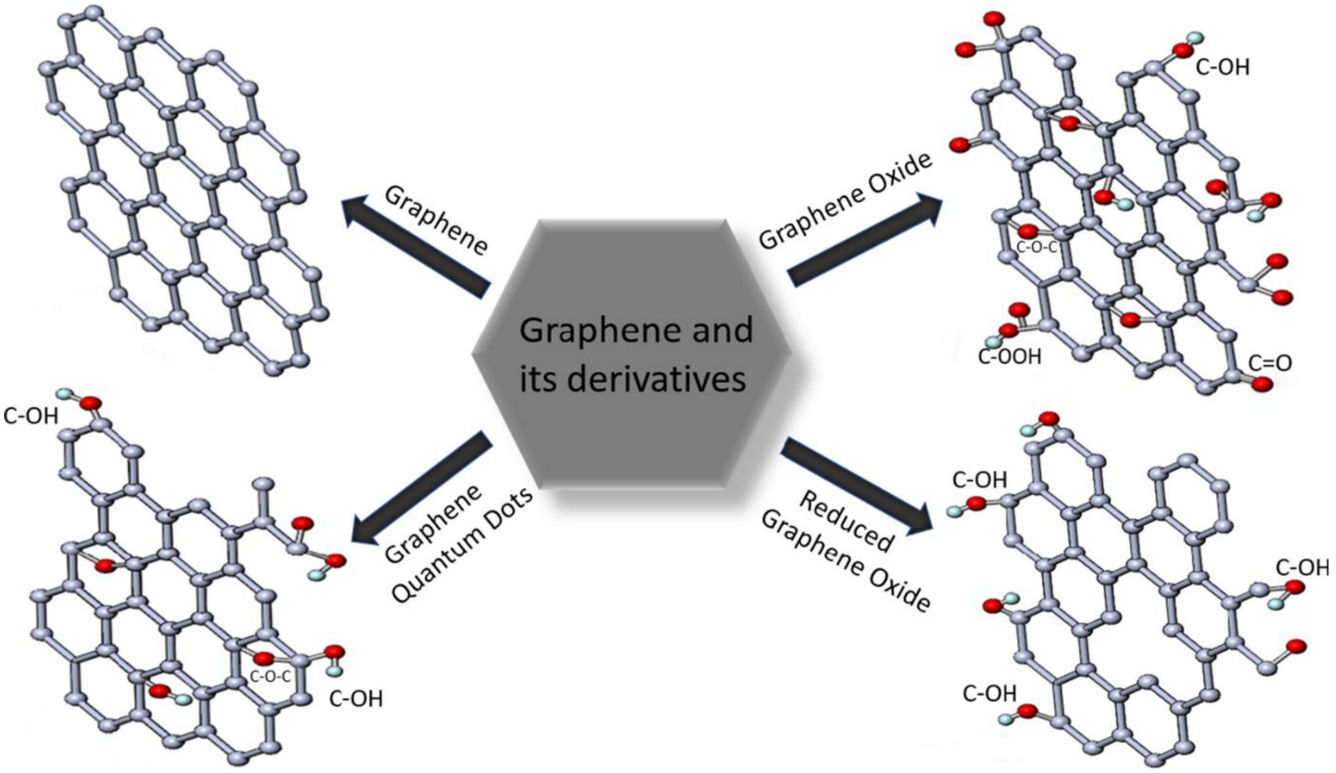


Figure 1. Graphene and its derivatives (reproduced with permission from [13]).

Pristine Graphene

Pristine graphene is the simplest form of graphene and consists of a single layer of carbon atoms arranged in a honeycomb lattice. This structure has exceptional properties, including high electrical conductivity [14], strength exceeding that of steel [15], and excellent thermal conductivity [16]. These traits make it ideal for electronic devices [17] and composite materials [18]. However, its large-scale use is limited by production challenges and difficulties in processing it in its pure form.

Graphene Oxide

GO is a graphene derivative with oxygen-containing functional groups such as hydroxyl, carboxyl, and epoxy groups, which disrupt its lattice and alter its properties [19]. These groups make GO more hydrophilic than pristine graphene, allowing it to disperse easily in water, which is ideal for liquid processing [20]. However, they also reduce the electrical conductivity of the material [21]. Despite this, GO is widely used in applications such as water purification [22] and as a precursor for other graphene-based materials such as rGO [23].

Reduced Graphene Oxide

rGO is produced by partially reducing GO [24] to restore some of the sp^2 carbon network, improving its electrical conductivity while retaining some oxygen groups [25]. This balance between hydrophilicity and conductivity makes rGO useful for applications such as supercapacitors [26] and batteries [27]. However, the reduction process is often incomplete, resulting in defects that affect its performance [28] and distinguish it from both pristine graphene and GO.

Graphene Quantum Dots (GQDs)

GQDs are nanoscale graphene fragments under 10 nanometers in size with unique electronic, optical, and chemical properties due to quantum confinement. Their tunable bandgap [29] makes them ideal for optoelectronics [30], bioimaging [31], sensing [32], and energy conversion [33]. Doping with elements such as nitrogen or sulfur further enhances their functionality [34].

1.1.2. Synthesis Methods for the Graphene-Based Materials

The synthesis of graphene and its derivatives is a critical aspect of their development, as the chosen method affects the material's properties and determines its suitability for different applications. Several synthesis methods have been developed (Figure 2), each with its own set of advantages, limitations, and areas of application.

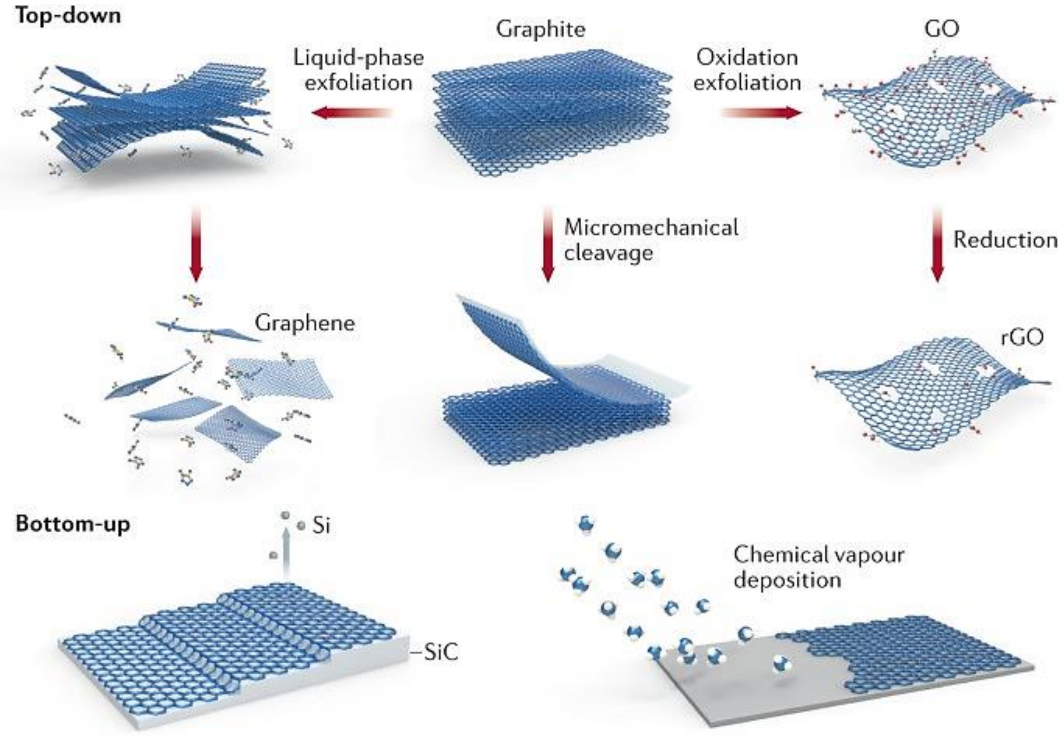


Figure 2. Different synthesis methods for graphene-based materials (reproduced with permission from [35]).

Liquid-Phase Exfoliation

Liquid-phase exfoliation (LPE) is a common method for synthesizing graphene from bulk graphite [36]. It involves dispersing graphite in a liquid medium with surfactants to prevent reaggregation. The process uses solvents such as N-methyl-2-pyrrolidone (NMP) [37] and applies ultrasonication to separate the graphene layers [38]. After sonication, exfoliated sheets are isolated from unexfoliated graphite through centrifugation [39]. While LPE is simple and scalable for large-scale production [40], it can introduce defects, contamination from surfactants, and variation in the thickness of the resulting graphene [41].

Oxidation Exfoliation for Graphene Oxide (GO) and Reduction for Reduced Graphene Oxide (rGO)

Oxidation exfoliation is a chemical method used to produce GO from graphite, which can be reduced to create rGO. The process begins by oxidizing graphite with strong agents such as sulfuric acid, nitric acid, and potassium permanganate [42], introducing oxygen-containing functional groups that increase the interlayer spacing for easier separation [43]. The oxidized graphite is then exfoliated in water to yield single- or few-layer GO, which can be reduced chemically [44], thermally [45], or electrochemically [46] to form rGO. While rGO has properties closer to those of pristine graphene, it still contains defects and residual oxygen groups [47]. This method is advantageous because of the high dispersibility of GO in solvents [48] and scalability [49] for producing graphene-based materials, although it has limitations such as lower electrical conductivity and environmental concerns related to the use of strong chemicals [50].

Micromechanical Cleavage

Micromechanical cleavage, or the “Scotch tape method”, is the original technique for isolating graphene from graphite and remains essential in graphene research [51]. This involves physically exfoliating graphene layers via adhesive tape, which is pressed onto a graphite crystal and then peeled off to remove thin layers. This process is repeated, and the tape is then pressed onto a substrate such as silicon dioxide (SiO_2) to transfer the graphene [52]. While this method produces high-quality, defect-free graphene that is ideal for research, it is not suitable for industrial-scale production because of its labor-intensive nature, low yield, and limited capacity for large quantities [53].

Silicon Carbide (SiC) to Graphene

The SiC-to-graphene method involves thermally decomposing SiC to produce high-quality, large-area graphene on its surface [54], making it suitable for electronic applications. The process starts with heating SiC in an ultrahigh vacuum or argon atmosphere [55], causing silicon atoms to sublimate and leave behind a carbon-rich surface that forms graphene layers. The number of layers can be controlled by adjusting the heating duration and temperature [56]. This method produces high-quality graphene with few defects, which is crucial for electronic applications [57], but it is expensive because of the cost of SiC substrates and high-temperature processing, and it can be challenging to uniformly control the number of layers over large areas [58].

Chemical Vapor Deposition (CVD)

CVD is a widely used method for producing large-area graphene films, particularly on metal substrates such as copper [59] or nickel [60]. The process starts by preparing and annealing the metal substrate to reduce defects. Hydrocarbon gases, such as methane [61], are then introduced at high temperatures and decompose on the surface to release carbon atoms that form graphene layers. After formation, the graphene can be transferred to other substrates by etching away the metal [62]. The main advantage of CVD is its ability to create continuous graphene films suitable for electronics, sensors, and transparent conductive films [63], although the transfer process can introduce defects or residues that affect quality [64].

2. Antibiotics in Wastewater: A Growing Environmental Concern

The presence of antibiotics in wastewater has emerged as a critical environmental issue, reflecting the unintended consequences of their widespread use and disposal [65]. Antibiotics, which were initially developed as life-saving drugs, have become pervasive in various sectors, including human [66] and veterinary medicine [67], agriculture [68], and aquaculture [69]. These compounds are now frequently detected in wastewater, raising alarms due to their potential impacts on ecosystems and public health [70]. The persistence of antibiotics in the environment, coupled with the inefficiencies of current wastewater treatment technologies, has resulted in their accumulation in natural water bodies, leading to serious ecological and health-related challenges [71]. To address this problem effectively, it is essential to understand the sources and environmental impacts related to antibiotics in wastewater.

2.1. Sources of Antibiotics in Wastewater

The entry of antibiotics into wastewater systems is multifaceted, stemming from various human activities and industrial processes (Figure 3). These sources contribute significantly to the contamination of water resources, making it a pressing environmental concern.

Figure 3. Different sources of antibiotics in wastewater.

2.1.1. Human Medicine

The most significant source of antibiotics in wastewater is human medicine [72]. After administration, a considerable portion of antibiotics consumed by humans are excreted unmetabolized. These excreted antibiotics enter the sewage system through urine and feces and are subsequently conveyed to wastewater treatment plants (WWTPs). Additionally, hospitals and healthcare facilities contribute heavily to this problem. They discharge not only antibiotics excreted by patients but also large quantities of expired or unused medications. In many cases, these drugs are disposed of directly into the sewage system, further increasing the concentration of antibiotics in wastewater [73]. The widespread use of antibiotics in outpatient settings, often without complete courses of treatment, exacerbates this issue by leading to more frequent and varied releases of these compounds into the environment.

2.1.2. Veterinary Medicine and Agriculture

The use of antibiotics in agriculture and animal husbandry is another major contributor to the presence of these drugs in wastewater [74]. In livestock farming, antibiotics are used not only to treat and prevent diseases but also to promote growth. Animals excrete a significant proportion of these antibiotics, which then enter the environment through manure and urine. When manure is used as fertilizer, antibiotics can leach into the soil and subsequently reach groundwater or be carried by runoff into surface water bodies [75]. In aquaculture, antibiotics are used to prevent infections in densely populated fish farms. These drugs are often administered through feed, and excess antibiotics, along with fish excreta, are directly discharged into surrounding water bodies, contributing to the contamination of aquatic environments [76]. The cumulative effect of these practices results in substantial antibiotic loads in the environment, which can eventually make their way into wastewater systems.

2.1.3. Pharmaceutical Manufacturing

The pharmaceutical industry is another significant source of antibiotics in wastewater [77]. During the production of antibiotics, wastewater generated from manufacturing processes can contain high concentrations of these drugs. In regions where environmental regulations are weak or poorly enforced, pharmaceutical plants may discharge their effluents directly into local water bodies or municipal sewage systems without adequate treatment [78]. This practice can lead to localized hotspots of antibiotic contamination, where the concentration of these drugs is much higher than that in typical domestic wastewater [79]. The environmental impact of these discharges is particularly concerning in developing countries, where regulatory oversight may be limited and industrial waste management practices are less stringent [80].

The presence of antibiotics in wastewater has profound implications for environmental health, primarily because of the promotion of antibiotic resistance [81], disruption of natural ecosystems [82], and potential risks to human health [83]. These impacts are complex and multifaceted, reflecting the intricate interplay between chemical pollutants and biological systems.

2.2. Properties of Graphene-Based Materials for the Treatment of Wastewater with Antibiotics

The unique properties of GBMs make them highly effective in the treatment of antibiotics in wastewater, addressing one of the most pressing environmental challenges. The extraordinary surface area of graphene, which is among the highest of any material, provides extensive active sites for the adsorption of antibiotic molecules, enabling efficient removal of these contaminants from water [84]. This high adsorption capacity, combined with the chemical stability of graphene [85], ensures that it can effectively capture and retain antibiotics, even in the presence of other competing contaminants. Furthermore, the exceptional electrical conductivity of graphene-based materials enhances their potential use in advanced oxidation processes (AOPs), where graphene can act as a catalyst or support material in the generation of reactive oxygen species (ROS) [86]. These ROS are capable of degrading antibiotics into less harmful compounds, thus reducing their environmental impact. The high thermal conductivity of graphene can also play a role in photothermal processes [87], where it absorbs light and converts it into heat, aiding in the degradation of antibiotic compounds under solar or artificial light. Moreover, the tunable surface chemistry of graphene allows functionalization with specific groups that can target and degrade certain classes of antibiotics, further increasing the specificity and efficiency of the treatment process [88]. These properties collectively position GBMs as powerful tools for developing advanced, sustainable technologies for the effective removal of antibiotics from wastewater, thereby mitigating the risk of environmental contamination and the spread of antibiotic resistance.

3. Research Methods

3.1. Systematic Literature Review

This study conducted a systematic literature review following the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) guidelines [89], which are widely recognized for enhancing the transparency, completeness, and accuracy of systematic reviews and meta-analyses. The PRISMA methodology [90] outlines a structured approach for identifying documents involving sequential steps: identification, screening, and inclusion based on eligibility quality assessment.

3.1.1. Identification

The literature search was carried out via the Scopus database [91], which was selected for its extensive coverage of recent and reputable publications across numerous journals. The search string used was “TITLE (graphene AND wastewater AND antibiotic)”, which was restricted to the “Article title” field in Scopus. This search yielded an initial set of 21 documents.

3.1.2. Screening

The 21 identified documents were then subjected to further screening based on predefined criteria: document type (Article), source type (Journal), publication stage (Final), and language (English). The following search string was applied to refine the results:

“TITLE (graphene AND wastewater AND antibiotic) AND (LIMIT-TO (DOCTYPE, “ar”)) AND (LIMIT-TO (PUBSTAGE, “final”)) AND (LIMIT-TO (SRCTYPE, “j”)) AND (LIMIT-TO (LANGUAGE, “English”))”.

This screening process, which was conducted on 24 September 2024, narrowed the list to 19 journal articles for further examination.

3.1.3. Eligibility

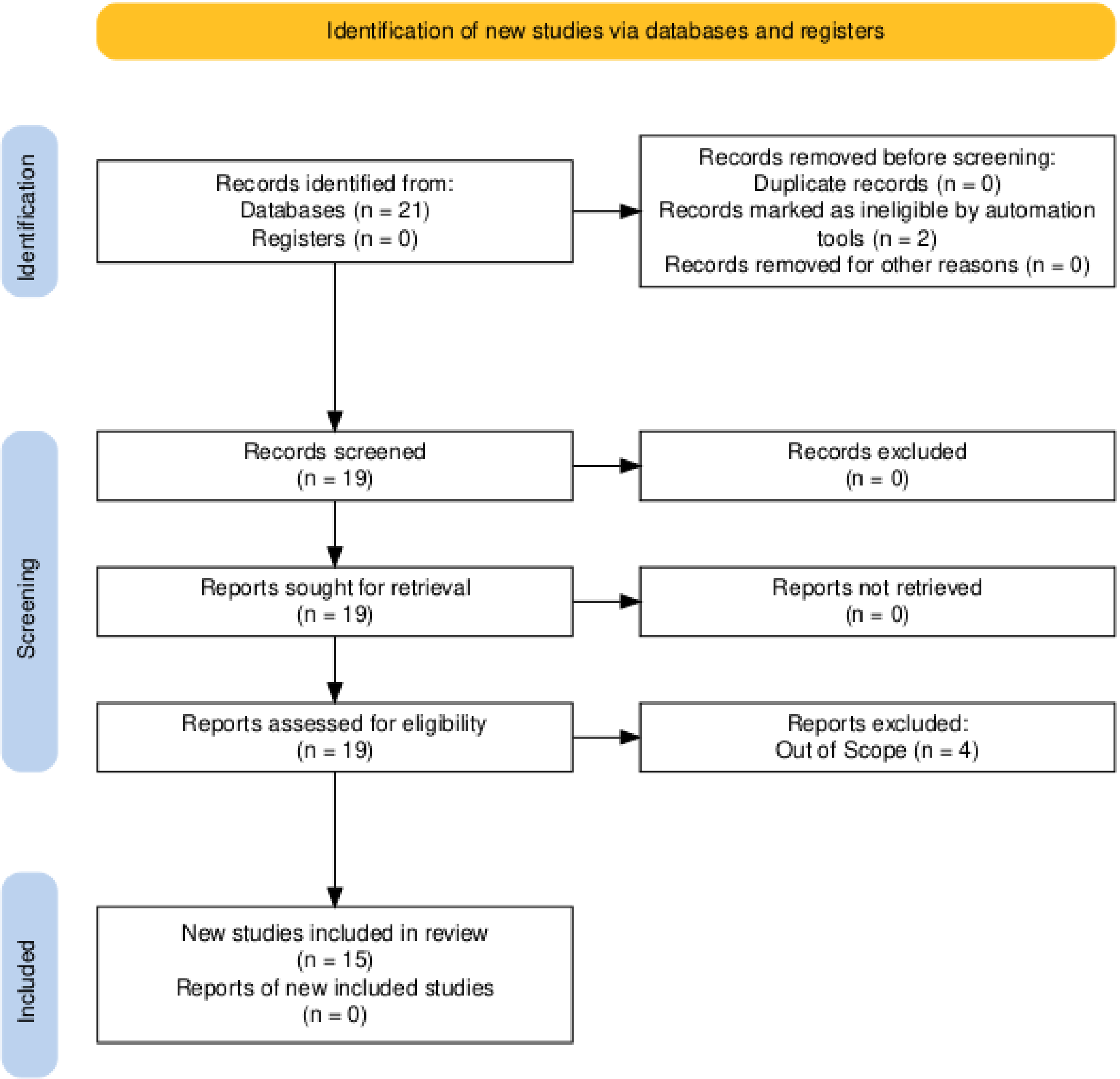
At the eligibility stage, the focus was on selecting journal articles specifically addressing graphene-based technologies for antibiotic removal from wastewater. The titles, abstracts, and full texts of the 19 articles were reviewed closely. Four articles were excluded because they were outside the scope of the study. Ultimately, 15 articles were deemed eligible for further analysis.

3.1.4. Included Articles and Quality Assessment

A total of 15 articles were retained for the final review. Bibliographic data, including author names, affiliations, article titles, keywords, abstracts, publication years, and journal titles, were extracted from the Scopus database and saved in a .csv file. A quality assessment of these articles was conducted via a checklist (Table 1). All 15 articles met the quality assessment criteria, ensuring their suitability for inclusion in the subsequent analysis and discussion (Scheme 1).

Table 1. Checklist of the quality assessment.

1. Are the aims and objectives explicitly articulated?
2. Is the reporting structured in a logical, cohesive, and coherent manner?
3. Is the proposed technique described in sufficient detail?
4. Is the research methodology aligned with the study's objectives?
5. Are the methods for data collection clearly and thoroughly explained?
6. Do the explanations and conclusions rely appropriately on the data presented?
7. Does the study make a meaningful contribution to the body of knowledge?
8. Has the stated aims and objectives been achieved?
9. Is the research process clearly and comprehensively documented?
10. Can the study be reproduced based on the information provided?



Scheme 1. Three steps of this systematic literature review following PRISMA, 2020 [92].

4. Removal of Antibiotics from Wastewater via Graphene-Based Materials

4.1. Removal of Antibiotics from Wastewater via Photocatalytic Degradation via Graphene-Based Materials

GBMs have demonstrated exceptional efficacy in the photocatalytic degradation of antibiotics from wastewater, leveraging their superior electronic properties and large surface area. These materials facilitate efficient removal processes, significantly reducing antibiotic concentrations and improving wastewater treatment outcomes.

The effectiveness of graphene-based TiO_2 composite photocatalysts for the degradation of the antibiotics sulfamethoxazole (SMX), erythromycin (ERY), and clarithromycin (CLA) under solar radiation in real urban wastewater was evaluated by Karaolia et al. [87]. The investigation involved synthesizing TiO_2–rGO composite photocatalysts via two ex-situ methods, hydrothermal and photocatalytic treatment, with GO and oxide P25 TiO_2 as precursor materials. This study aimed to compare the photocatalytic efficiency of synthesized TiO_2–rGO composites with that of pristine Aeroxide P25 TiO_2 for the removal of these antibiotics from urban wastewater effluents pretreated with a membrane bioreactor and subsequently exposed to simulated solar radiation. The findings revealed that the TiO_2–rGO composite prepared via photocatalytic treatment demonstrated superior photocatalytic degradation of ERY (84 ± 2%) and CLA (86 ± 5%), whereas the degradation of SMX (87 ± 4%) was slightly more effective with Aeroxide P25 TiO_2.

Yazdi et al. [93] investigated a method for synthesizing ternary nanocomposites consisting of TiO_2 nanosheets, rGO, and various amounts of Ag for the photocatalytic treatment of tetracycline (TC) antibiotic wastewater. Plasmonic Ag nanoparticles were incorporated into the TiO_2 nanosheet/rGO nanocomposite through a photo deposition technique (TGA(x) samples). The photocatalytic activity of these nanocomposites was assessed for the degradation of TC at an initial concentration of 30 mg/L, with the TGA (0.076) sample achieving a degradation efficiency of 52.56% after 3 h of visible light exposure. The study revealed that the rGO in the TGA(x) samples significantly facilitated the transfer of photoinduced electrons from the plasmonic Ag nanoparticles to the TiO_2 nanosheets. Additionally, a three-layer artificial neural network model incorporating four input variables, such as irradiation time, catalyst dosage, initial TC concentration, and silver nitrate content, and an output variable, i.e., % degradation, was optimized with 11 hidden neurons. Analysis via Garson's formula revealed that the initial TC concentration was the most influential factor affecting treatment efficiency, accounting for 31% of the relative importance.

In a recent investigation, Yang et al. [94] developed an advanced photocatalytic flow-through system that incorporated a three-dimensional (3D) photoanode along with a Pt/C air-breathing electrode as the cathode (Figure 4). The system's efficacy in degrading ampicillin was evaluated by comparing the photocatalytic performance of a C_3N_4-MoS_2 composite supported on a 3D graphene matrix (C_3N_4-MoS_2/3DG) with that of other 3D materials. The findings revealed that the photoanode achieved a 74.6% removal efficiency for ampicillin within 2 h of the reaction. Subsequent toxicity evaluations, including acute antibacterial potency assessments and ECOSAR model predictions, indicated a reduction in ampicillin toxicity following photocatalytic treatment, likely due to the cleavage of functional groups such as amido and peptide bonds. The superior performance of the flow-through system was attributed to the enhanced adsorption capacity and heterojunction mechanism of the C_3N_4-MoS_2/3DG composite, along with the optimized mass and electron transfer characteristics inherent to the flow-through design. Additionally, the synergistic interaction between the photoanode and cathode was identified as a significant factor contributing to the system's overall efficiency. Consequently, this photoanode-based flow-through cell system is recognized as a promising strategy for the effective removal of organic pollutants in wastewater treatment processes.

In their research, Ivan et al. [22] synthesized hybrid layers composed of Fe oxide, Fe hydroxide, and nitrogen-doped graphene-like platelets through an eco-friendly laser-based method aimed at photocatalytic applications. These composite layers exhibited considerable efficiency in the photodecomposition of antibiotic molecules under visible light irradiation. The efficiency of photodecomposition was systematically analyzed in relation to varying concentrations of base materials, including the Fe oxide nanoparticles and GO platelets used in dispersions subjected to laser irradiation. Although the pure Fe oxide/Fe hydroxide layers strongly absorbed visible light, their photodecomposition efficiency under comparable conditions was negligible. The improved photocatalytic efficiency of the nanohybrid layer, which achieved up to 80% degradation of the initial antibiotic molecules, was attributed to synergistic interactions among the composite materials. These interactions facilitated the effective separation of electron–hole pairs generated on the surfaces of Fe oxide and Fe hydroxide nanoparticles under visible light, with conductive graphene-like platelets playing a critical role. Furthermore, nitrogen-doped graphene-like platelets contributed to the generation of electron–hole pairs under visible light, as demonstrated by the photocatalytic activity observed in the pure, nitrogen-doped graphene-like reference layers. The study also indicated that adsorption processes played a minimal role in the removal of antibiotic molecules from the test solutions, with the observed reduction in antibiotic concentration primarily resulting from photocatalytic decomposition mechanisms.

Lin et al. [95] developed a GO structure-oriented NH_2-MIL-88B(Fe)/GO/sodium alginate (NM88B/GO/SA) aerogel for antibiotic removal (Figure 5). The aerogel was fabricated via GO interface regulation and dual-network crosslinking techniques, resulting in the formation of an NM88B/GO heterostructure via the directional growth of NM88B on the GO surface. Sodium alginate (SA) was incorporated to increase the robustness of the matrix and prevent fragmentation. With a 30 wt% catalyst loading, the composite aerogel demonstrated exceptional photocatalytic performance, achieving over 99% removal of high-concentration (50 ppm) tetracycline hydrochloride (TC-HCl) within 150 min. The aerogel also exhibited notable shape recovery, stability, and reusability, maintaining over 95% degradation efficiency for TC-HCl after five repeated tests. This aerogel has considerable potential as an efficient and reusable photocatalyst for wastewater treatment.

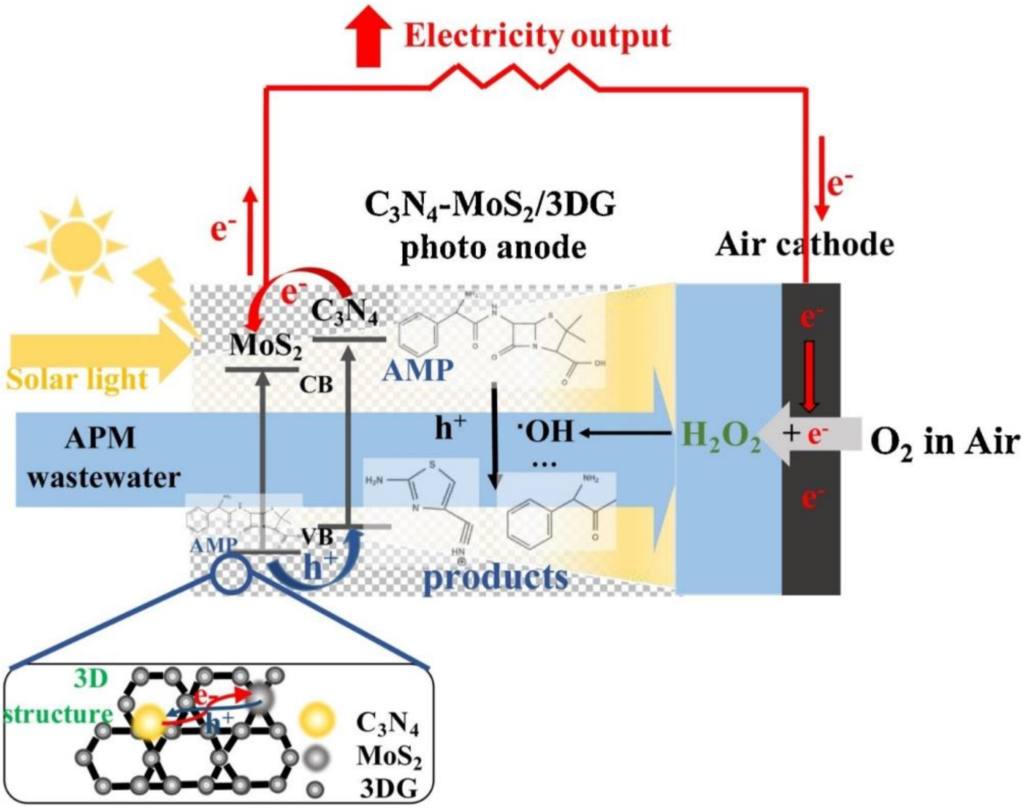


Figure 4. Diagram depicting AMP wastewater treatment via the C_3N_4-MoS_2/3DG flow-through system, enhanced by the addition of trace amounts of H_2O_2 and electricity generation by the air cathode. (Reproduced with permission from [94]).

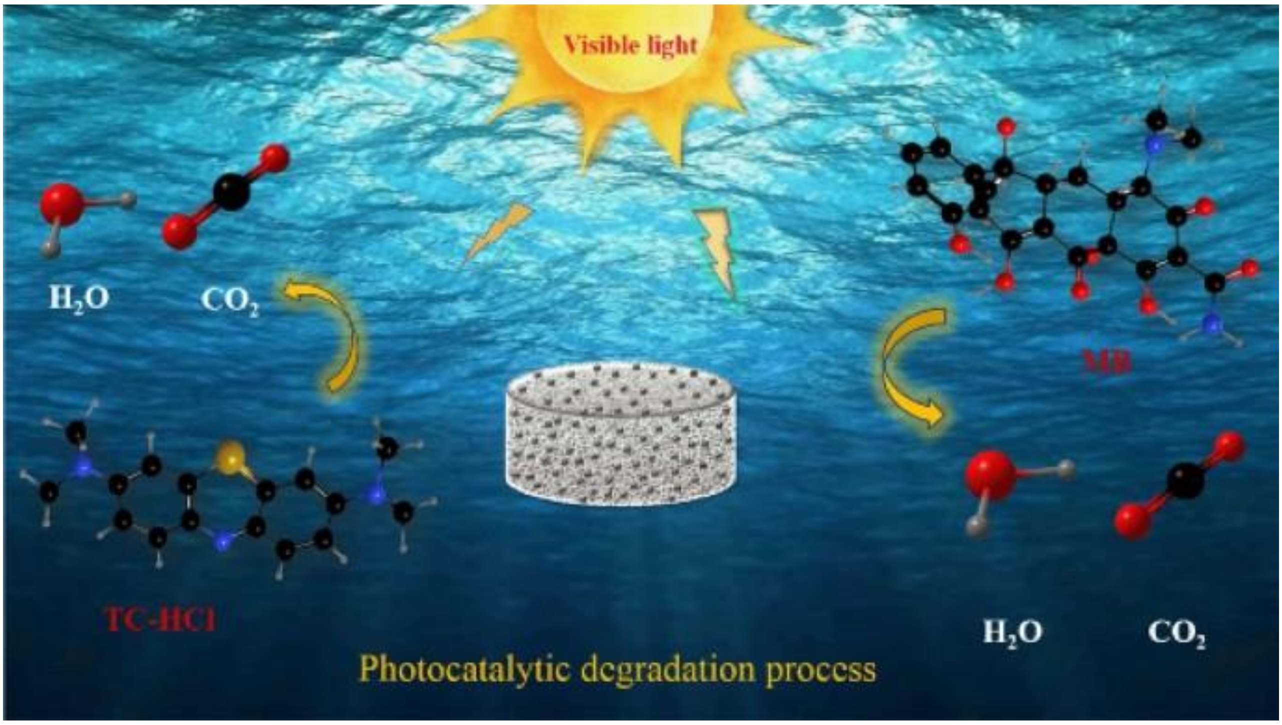


Figure 5. GO-based NM88B/GO/SA aerogels for antibiotic-contaminated wastewater (reproduced with permission from [95]).

4.2. Removal of Antibiotics from Wastewater via Adsorption via Graphene-Based Materials

GBMs have demonstrated exceptional efficacy in the adsorption of antibiotics from wastewater, significantly reducing their concentration. The high surface area and tunable properties of these materials enhance their removal efficiency, making them promising candidates for advanced water treatment technologies.

A one-pot solvothermal synthesis method was employed by Wu et al. [96] to produce RGO-supported ferrite hybrids via the use of GO and Fe^{3+} ions as precursors. The resulting Fe_3O_4 nanoparticles were uniformly dispersed and anchored onto the RGO nanosheets. These nanocomposites exhibited high adsorption efficiency for quantifying three sulfonamides (SAs) in wastewater, which was attributed to the large surface area of the RGO. Several key experimental parameters, including the amount of adsorbent, extraction time, pH, and desorption conditions, were systematically optimized to increase the extraction efficiency. Compared with conventional adsorbents, the RGO–Fe_3O_4 nanocomposites demonstrated superior extraction capabilities. A broad linear detection range of 1 to 200 ng/mL was achieved, with a correlation coefficient above 0.9987, and the detection limits for the three sulfonamides ranged between 0.43 and 0.57 ng/mL. When applied to environmental wastewater samples, the method achieved recoveries ranging from 89.1% to 101.7%, with relative standard deviations below 8.6%, confirming its efficacy and reproducibility across different matrices.

Radmehr et al. [84] synthesized a renewable adsorbent, NiZrAl-layered double hydroxide-graphene oxide-chitosan (NiZrAl-LDH-GO-CS NC), for the effective removal of nalidixic acid (NA) from wastewater. The successful synthesis was confirmed through various characterization techniques. The NiZrAl-LDH-GO-CS NCs exhibited rapid adsorption rates and a maximum adsorption capacity of 277.79 mg/g, surpassing those of other nanobased adsorbents. The adsorption process was analyzed via a central composite design, adaptive network-based fuzzy inference system, and general regression neural network models, with the adaptive network-based fuzzy inference system showing superior performance in predicting NA removal. Optimization through response surface methodology (RSM) achieved over 92% NA removal. Thermodynamic and kinetic assessments revealed chemisorption mechanisms. The study concluded that the LDH-GO-CS adsorbent has significant potential for pharmaceutical wastewater treatment and warrants further research for practical remediation applications.

In their study, Behzadi et al. [97] investigated the use of organic aerogels with high specific surface areas for the removal of TC antibiotics. They synthesized resorcinol formaldehyde (RF) aerogels via a sol-gel process, followed by drying under ambient conditions. To enhance the adsorbent properties, they modified RF aerogels with 1 wt.% graphene and 1 wt.% m-phenylenediamine, producing the RF-G1/PmPDA1 composite. The performance of these modified aerogels was assessed under various conditions, including different pH values (ranging from 2–12), adsorbent doses (4–10 mg), and contact times with antibiotics (3–24 h). Brunauer–Emmett–Teller (BET) analysis indicated that the modification increased the specific surface area of the RF aerogel from 96 to 308 m^2/g. The RF aerogel and RF-G1/PmPDA1 composites achieved TC removal rates of 65.2% and 93.3%, respectively, at an optimal pH of 4.

Kogut et al. [98] evaluated the feasibility of using adsorptive nonwovens as a cost-effective pretreatment strategy in wastewater treatment, highlighting their potential for environmental protection through the efficient adsorption of antibiotics. Research has concentrated on graphene-modified nonwovens (GMNs) and investigated two primary aspects: the adsorption behavior of TC, a common environmental contaminant at concentrations relevant to real-world conditions, and the factors influencing the antibacterial and antifungal properties of these materials. The study revealed that the integration of graphene particles with commercial textile auxiliaries significantly enhanced the antibacterial and antifungal properties of the nonwovens. TC residues were detected at the ng/mL scale via enzyme-linked immunosorbent assay (ELISA), and the adsorption behavior followed Henry and Redlich–Peterson isotherms, demonstrating the effectiveness of the process at low TC concentrations. As a result, well-designed GMNs have emerged as promising candidates for wastewater treatment applications, particularly in sewage treatment facilities. Additionally, statistical analyses of nonwoven and modified nonwoven morphologies, including skewness and kurtosis, provided valuable insights into the parameters influencing fungal growth in these structures. The research concluded that the GMNs exhibited significant antibiotic adsorption capabilities, resulting in a two-fold reduction in the TC concentration during the experiments.

Rajapaksha et al. [99] investigated the impact of pharmaceutical discharge on wastewater contamination, highlighting the significant environmental and public health risks posed by these pollutants. They synthesized GO by oxidizing graphite via a modified Hummers method, which was then used to develop two composite materials (Figure 6): copper oxide-reduced graphene oxide (CuO–rGO) and zinc oxide-reduced graphene oxide (ZnO–rGO). The adsorption capacities of these composites for the antibiotics amoxicillin (AMOX) and TC were evaluated via UV–visible spectroscopy. The CuO–rGO composite exhibited particularly high antibiotic removal efficiencies, achieving capacities of 405 mg/g for AMOX and 552 mg/g for TC at a pH of 7 and a temperature of 333 K. Additionally, this material demonstrated an 80% regeneration efficiency and retained 82% of its adsorption capacity after five reuse cycles. The adsorption behavior of AMOX and TC on GO, CuO–rGO, and ZnO–rGO followed the Langmuir isotherm and pseudo-second-order kinetic models, indicating a chemisorption process. Thermodynamic analyses revealed that these adsorption processes were spontaneous, with exothermic adsorption observed for AMOX and TC on CuO–rGO and ZnO–rGO, whereas adsorption on GO was endothermic. This study emphasized the enhanced efficacy of rGO materials in removing anionic antibiotics from water, suggesting that electrostatic interactions and π–π interactions between antibiotics and adsorbents are key mechanisms. The findings also suggested that, while unmodified GO was effective for the adsorption of cationic contaminants, the rGO composites were more suitable for targeting anionic contaminants in wastewater treatment applications.

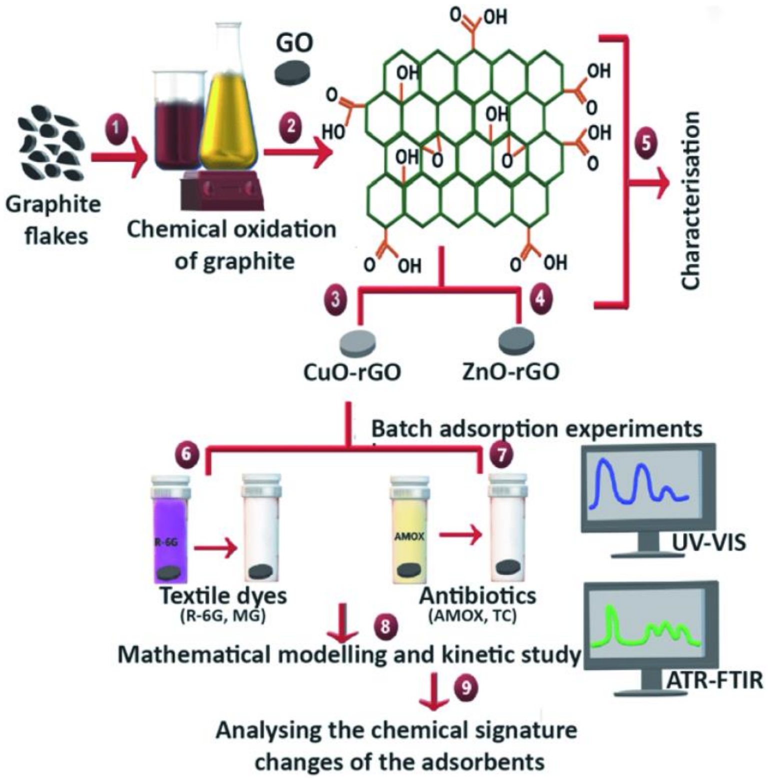


Figure 6. A schematic of the experimental design included the following steps: (1) Synthesizing graphene oxide (GO) from raw graphite flakes via oxidation via a modified Hummers method. (2) Obtaining the resulting GO. (3) Copper oxide-doped reduced graphene oxide (CuO–rGO) was synthesized from synthesized GO and an aqueous $CuSO_4 \cdot 5H_2O$ solution, and (4) zinc oxide-doped reduced graphene oxide (ZnO–rGO) was synthesized from synthesized GO and an aqueous $ZnSO_4 \cdot 7H_2O$ solution through a series of thermal chemical reactions. (5) The chemical and physical properties of GO, CuO–rGO, and ZnO–rGO were characterized via standard microscopic and spectroscopic techniques, including SEM, TEM, ATR-FTIR, and XPS. (6) Batch adsorption experiments were conducted to remove textile dyes (rhodamine 6G (R-6G) and malachite green (MG)) and antibiotics (amoxicillin (AMOX) and tetracycline (TC)) from aqueous solutions via GO, CuO–rGO, and ZnO–rGO adsorbents, followed by analysis via UV–visible spectroscopy. (7) Mathematical modeling and kinetics were applied to study the batch adsorption of textile dyes (R-6G, MG) and antibiotics (AMOX, TC) on GO, CuO–rGO, and ZnO–rGO. (8) Analyzing functional group changes on the GO, CuO–rGO, and ZnO–rGO adsorbents after adsorption of the textile dyes and antibiotics via ATR-FTIR. (Reproduced with permission from [99]).

Taleb et al. [100] addressed the pressing issue of antibiotic removal from contaminated water bodies, which is a critical step toward sustainable development. They synthesized a multifunctional hybrid thin film composed of carboxymethyl cellulose, GO, and polyaniline (CMC/GO/PANI) and conducted a systematic evaluation of its effectiveness in adsorbing oxytetracycline (OTC) from wastewater. The adsorption process was investigated under various conditions, including different reaction times, pH values, concentrations, and temperatures. The findings revealed that the CMC/GO/PANI hybrid thin film exhibited better OTC adsorption than the individual CMC, GO/CMC, and PANI/CMC thin films, which was attributed to the synergistic interaction of its multifunctional components. The adsorption kinetics were best described by the pseudo-second-order model, whereas the Redlich–Peterson isotherm provided an accurate interpretation of the adsorption equilibrium. Thermodynamic analysis indicated that the process was both spontaneous and endothermic. Moreover, the CMC/GO/PANI thin film demonstrated significant reusability, maintaining its adsorption capacity across seven cycles of adsorption and desorption. This study underscores the substantial potential of the CMC/GO/PANI thin film for large-scale wastewater purification, highlighting its durability and enhanced adsorption performance.

AbuZaid et al. [101] investigated the problem of elevated antibiotic levels in wastewater, which exacerbated the emergence of antimicrobial-resistant pathogens and intensified global water scarcity, adversely affecting both environmental systems and human health. Their research focused on the synthesis and assessment of a two-dimensional (2D) lamellar, free-standing GO/Ti_3C_2Tx membrane designed for the effective removal of antibiotics from wastewater (Figure 7). The 50% GO/Ti_3C_2Tx composite membrane significantly increased the water flux, reaching 61.9 L m^{-2} h^{-1}, which markedly exceeded the flux of the pristine GO membrane at 22.8 L m^{-2} h^{-1}. Moreover, the GO/Ti_3C_2Tx composite membranes achieved TC rejection rates consistently above 99%, representing a substantial improvement over the performance of the pristine Ti_3C_2Tx membrane. The membranes with GO to MXene ratios of 50%, 40%, 30%, and 20% were found to have contact angles of 54.5°, 57.4°, 60.6°, and 61.6°, respectively. The study highlighted that the integration of GO and Ti_3C_2Tx enhanced the membrane properties, as indicated by modifications in the interlayer spacing and hydrophilicity. Additionally, compared with both the unmodified GO and Ti_3C_2Tx membranes, the composite membrane exhibited superior antifouling properties.

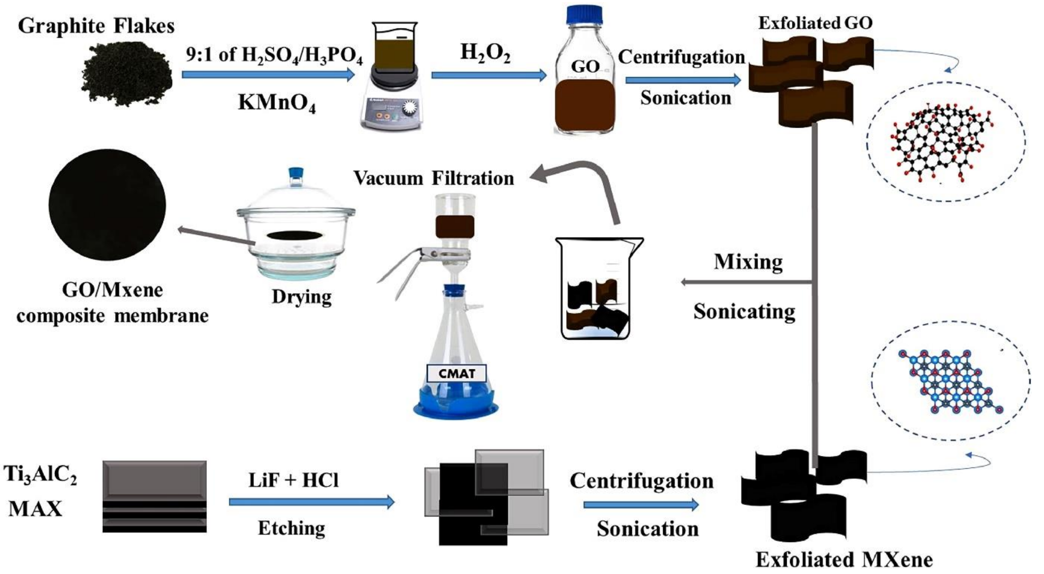


Figure 7. Diagrammatic representation of the synthesis process for free-standing graphene oxide (GO), Ti_3C_2Tx, and GO/Ti_3C_2Tx composite membranes (reproduced with permission from [101]).

Shaker et al. [102] employed a nanocomposite composed of copper nanoparticles immobilized on polyaniline-modified GO (Cu/PANI/GO) to successfully extract TC from environmental water samples. Compared with unmodified GO, the Cu/PANI/GO nanocomposite substantially improved the TC removal efficiency, achieving a remarkable removal rate of 99.6% and an adsorption capacity of 434.78 mg/g. This enhanced performance was attributed to the synergistic effects between the copper nanoparticles and polyaniline components, which led to a TC removal efficiency of approximately 51.5 ± 3.9% greater than that observed with GO alone (Figure 8). The adsorption kinetics and isotherms of TC on the Cu/PANI/GO nanocomposite were accurately modeled via the pseudo-second-order kinetic model and the Langmuir isotherm model, respectively. Additionally, the nanocomposite demonstrated notable durability over five adsorption–desorption cycles, with only a 4.1% reduction in removal efficiency. Researchers have proposed that the strong retention of TC within the Cu/PANI/GO structure results from surface complexation by copper nanoparticles, effective hydrogen bonding with polyaniline and GO, and π–π interactions with GO. Thus, the Cu/PANI/GO nanocomposite proved to be an effective material for the removal of TCs from wastewater samples.

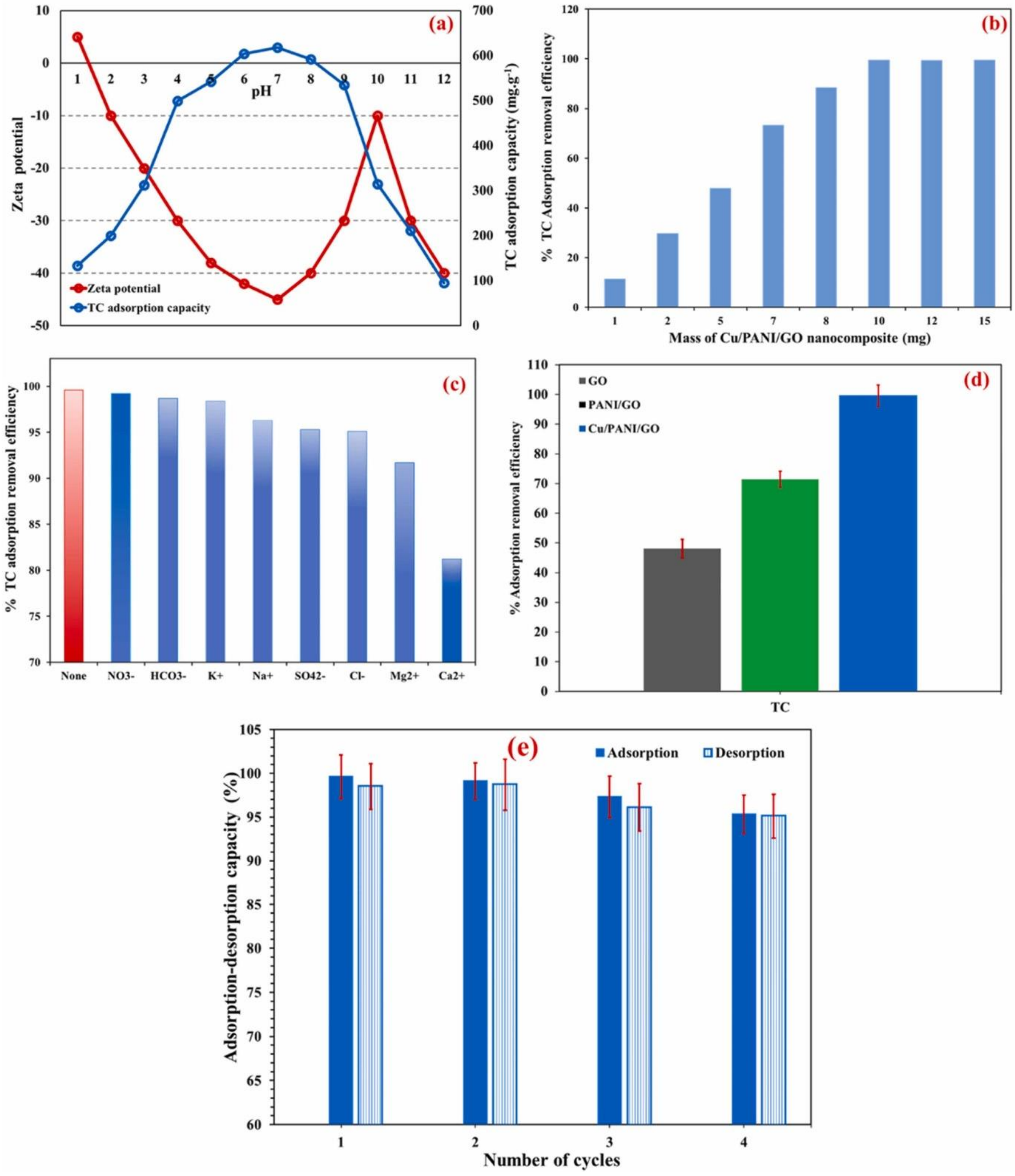


Figure 8. Influence of various factors on the tetracycline adsorption efficiency of the Cu/PANI/GO nanocomposite: (**a**) pH and zeta potential, (**b**) mass dosage (mg), (**c**) presence of interfering ions, (**d**) synergistic impact of Cu nanoparticles and polyaniline on the adsorption efficiency, and (**e**) reusability of the Cu/PANI/GO nanocomposite over four consecutive cycles under optimal conditions (reproduced with permission from [102]).

4.3. Removal of Antibiotics from Wastewater via Other Methods via Graphene-Based Materials

There are alternative methods, in addition to photocatalytic degradation and adsorption, for removing antibiotics from wastewater.

Solís et al. [88] developed magnetic graphene-based catalysts aimed at removing a variety of antibiotics, including SMX, norfloxacin, TC, and flumequine, from water (Figure 9). Their study investigated the catalytic activation of inorganic peroxides, namely, peroxymonosulfate (PMS), peroxydisulfate, and hydrogen peroxide, by altering the ratio of magnetite to graphene from 1:0 to 0:1. The results indicated that graphene played a crucial role in catalytic activation, with the process being most efficient in the presence of PMS. A magnetite concentration of 20% in the solid matrix was adequate for the complete degradation of the antibiotics, and the use of a magnetic field facilitated effective recovery. The performance of the catalyst was further evaluated in simulated urban wastewater, where key factors influencing the process and its stability over multiple reuse cycles were examined. Kinetic scavenging probe tests corroborated the hypothesis of a non-radical mechanism during PMS activation, whereas electron paramagnetic resonance analysis in the presence of D_2O revealed that electron transfer was the predominant reaction mechanism. The magnetic catalyst demonstrated both high catalytic activity and stability, proving to be an effective method for removing antibiotics from water.

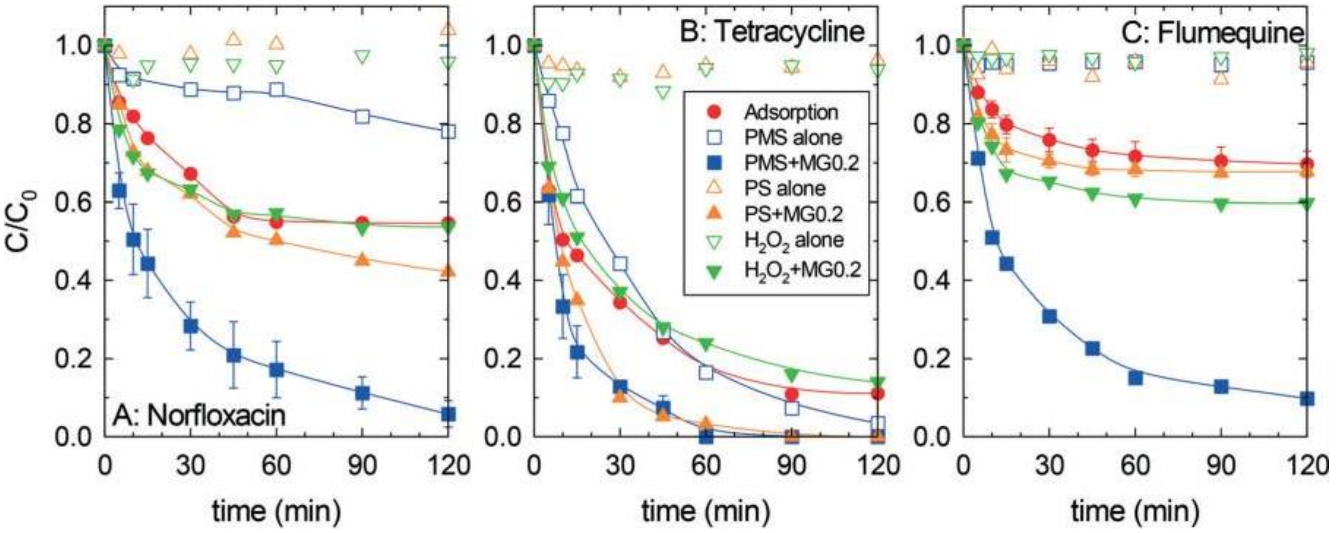


Figure 9. Removal of norfloxacin (**A**), tetracycline (**B**), and flumequine (**C**) via activated inorganic peroxides with magnetic graphene MG0.2 (reproduced with permission from [88]).

Peng et al. [86] developed a flow-through electrochemical system that utilized a series of graphene nanoparticle-loaded PbO_2 reactive electrochemical membrane electrodes (GNP-PbO_2 REMs) mounted on porous titanium substrates with different pore sizes for the treatment of antibiotic-contaminated wastewater. Among these, the GNP-PbO_2 electrodes supported on titanium substrates with a pore size of 150 μm (Ti-150/GNP-PbO_2) exhibited superior electrochemical degradation performance compared with REMs with other pore sizes. This improved performance was attributed to the smaller crystal size, larger electrochemically active surface area, reduced charge-transfer impedance, and higher oxygen evolution potential of Ti-150/GNP-PbO2. Under optimized conditions with an initial pH of 5, the Ti-150/GNP-PbO_2 REM achieved a benzylpenicillin sodium (PNG) removal rate of 99.34%. This performance was attributed to enhanced mass transfer. This study proposed three plausible degradation pathways for PNG within a flow-through electrochemical system and confirmed the stability and safety of the Ti-150/GNP-PbO_2 REM.

A prior discussion on the removal of antibiotics from wastewater via GBMs is summarized in Table 2.

Table 2. Graphene-based materials are used for the removal of antibiotics from wastewater.

Graphene Type	Target	Type of Treatment	Results Achieved	Reference
Reduced Graphene Oxide	sulfamethoxazole (SMX), erythromycin (ERY), and clarithromycin (CLA)	Photocatalytic degradation	Degradation efficiency SMX (87 ± 4%), ERY (84 ± 2%), CLA (86 ± 5%)	Karaolia et al. [87]
Reduced Graphene Oxide	Tetracycline	Photocatalytic degradation	Degradation efficiency 52.56%	Yazdi et al. [93]
Graphene	Ampicillin	Photocatalytic degradation	Removal efficiency 74.6%	Yang et al. [94]
Reduced Graphene Oxide	Chloramphenicol sodium succinate	Photocatalytic degradation	Removal efficiency 80%	Ivan et al. [22]
Graphene Oxide	Tetracycline	Photocatalytic degradation	Removal efficiency > 99%	Lin et al. [95]
Reduced Graphene Oxide	Sulfonamide	Adsorption	Recoveries range from 89.1 and 101.7%	Wu et al. [96]
Graphene Oxide	Nalidixic acid	Adsorption	Adsorption capacity 277.79 mg/g (Removal efficiency 92%)	Radmehr et al. [84]
Graphene	Tetracycline	Adsorption	Removal efficiency 93.3%	Behzadi et al. [97]
Graphene	Tetracycline	Adsorption	NA	Kogut et al. [98]
Graphene Oxide	Amoxicillin and Tetracycline	Adsorption	Adsorption capacity 405 mg/g and 552 mg/g for Amoxicillin and Tetracycline, respectively	Rajapaksha et al. [99]
Graphene Oxide	Oxytetracycline	Adsorption	Adsorption capacity 180.240 mg/g (Removal efficiency > 90%)	Taleb et al. [100]
Graphene Oxide	Tetracycline	Adsorption	Removal efficiency 99.8%	AbuZaid et al. [101]
Graphene Oxide	Tetracycline	Adsorption	Adsorption capacity 434.78 mg/g (Removal efficiency 99.6%)	Shaker et al. [102]
Graphene	Sulfamethoxazole, Norfloxacin, Tetracycline, Flumequine	Catalytic activation of inorganic peroxides	NA	Solís et al. [88]
Graphene	Benzylpenicillin sodium	Electrochemical oxidation	Removal efficiency 99.34%	Peng et al. [86]

Thus, GBMs play a pivotal role in removing antibiotics from wastewater through various mechanisms, including photocatalytic degradation, adsorption, catalytic activation of inorganic peroxides, and electrochemical oxidation. In photocatalysis, graphene enhances the efficiency of semiconductor catalysts by improving charge separation and increasing the generation of ROS, which degrade antibiotics. Its high surface area and functional groups enable strong adsorption of antibiotic molecules, facilitating their removal. Additionally, graphene acts as a catalyst in the activation of inorganic peroxides, generating ROS through Fenton-like reactions that break down antibiotics. In electrochemical oxidation, the excellent conductivity of graphene promotes direct electron transfer and ROS formation, further enhancing antibiotic degradation. These diverse functions make graphene materials highly effective for antibiotic removal, although challenges remain in scaling their use for practical applications.

Antibiotic removal from wastewater is a critical environmental challenge, and two promising approaches, adsorption, and photocatalysis, leverage the unique properties of graphene to address this issue effectively. Adsorption offers a straightforward method for removing antibiotics by capturing contaminants on the graphene surface, benefiting from the high surface area and tunable chemical properties of functionalized graphene. This method is highly effective, especially for a wide range of antibiotics, and allows for simple operation without requiring light sources or complex equipment. However, adsorption has limitations, such as potential saturation of the material, which reduces its effectiveness over time and necessitates regeneration processes that may degrade the material's performance.

Photocatalysis, on the other hand, introduces an additional advantage by degrading antibiotics rather than merely trapping them, breaking them down into less harmful substances using light energy. Graphene combined with metal oxides, like TiO_2 or ZnO, enhances this process due to improved electron transfer, which increases photocatalytic efficiency. Despite this, photocatalysis requires external energy input (light) and specific conditions to function optimally, which can limit its application in large-scale or low-resource settings. Moreover, the potential release of toxic byproducts during degradation is a concern. Thus, while both methods offer significant potential for antibiotic removal, their practical implementation should carefully consider these performance trade-offs.

5. Challenges, Potential Solutions and Mitigation Strategies

The application of advanced graphene-based technologies for antibiotic removal from wastewater faces several challenges. Graphene materials, despite their remarkable properties, often face issues related to scalability [103] and cost-effectiveness [104]. The synthesis and functionalization of GBMs can be complex and expensive, limiting their widespread adoption [105]. Additionally, the removal efficiency of these technologies can be influenced by the presence of other contaminants and the variability in wastewater composition. The effectiveness of graphene-based systems in degrading or adsorbing antibiotics may vary depending on the specific antibiotic and its chemical properties, as well as the operational conditions [106]. Furthermore, potential concerns regarding the stability and longevity of graphene materials in real-world wastewater treatment scenarios need to be addressed to ensure their practical applicability [107].

To address these challenges, research into optimizing the synthesis and functionalization processes of graphene-based materials is essential. The development of cost-effective and scalable methods for producing graphene and its composites could increase their viability for large-scale wastewater treatment applications [108]. Advanced techniques, such as doping [109] or facile functionalization [110], can be employed to tailor graphene materials for improved performance in removing specific antibiotics. Additionally, integrating graphene-based technologies with other advanced treatment methods, such as electrochemical processes [86], could increase their efficiency and broaden their applicability. Collaborative research efforts and innovations in materials science are crucial for overcoming current limitations and maximizing the potential of graphene-based technologies in wastewater treatment.

Effective strategies for overcoming the challenges associated with graphene-based technologies involve a multipronged approach. Regulatory support and funding for research into graphene-based water treatment technologies can accelerate the development of cost-effective solutions [111]. Public and private sector collaborations can facilitate the commercialization of these technologies by addressing scale-up challenges and integrating them into existing treatment infrastructure. Additionally, ongoing research should focus on developing robust methods for evaluating the performance and stability of graphene-based systems in diverse wastewater conditions. Implementing pilot-scale studies and real-world applications will provide valuable insights into the practical challenges and performance of these technologies, guiding future improvements and ensuring their successful deployment in wastewater treatment systems.

GBMs have demonstrated remarkable efficacy in removing antibiotics from wastewater, offering a promising solution to this pressing environmental issue [112]. However, given the current regulatory landscape, specifically the maximum allowable concentration (MAC) values for antibiotics in wastewater, alternative methods that are more cost-effective and can still meet effluent quality standards are often preferred. According to environmental protection agencies, the MAC values for antibiotics in wastewater generally range between 0.01 and 1 μg/L, depending on the antibiotic type and the environmental impact [113].

While alternatives such as activated carbon [114] or biological treatments [115] may meet these regulatory requirements at a lower cost, graphene-based technologies are emerging as viable long-term solutions. Compared with traditional materials, the unique properties of graphene, such as a high surface area, excellent adsorptive capabilities, and the potential for functionalization, provide superior antibiotic removal efficiency [116].

The future direction of wastewater treatment could lean heavily towards graphene-based materials, especially as the costs of production decrease [117] and regulations potentially tighten to require more advanced removal techniques. As more research substantiates the long-term environmental safety and effectiveness of these materials [118], they may become the preferred choice for meeting stringent effluent standards. Furthermore, the multifunctionality of graphene, such as its use in catalytic degradation and its potential for regeneration, makes it a sustainable and scalable solution for antibiotic removal in wastewater [112].

GBMs adsorb antibiotics through various interaction mechanisms, which include π–π stacking [119], hydrogen bonding [120], electrostatic interactions [121], van der Waals forces [120], and hydrophobic interactions [120]. π–π stacking occurs between antibiotics with aromatic rings and the delocalized π–electron system of graphene. Hydrogen bonding forms between functional groups in antibiotics and oxygen-containing groups on graphene oxide, whereas electrostatic interactions depend on the charge of both antibiotics and functionalized graphene surfaces and are influenced by pH. Van der Waals forces contribute to the weaker, nonspecific adsorption and hydrophobic interactions that occur between the hydrophobic portions of antibiotics and the graphene surface. These diverse mechanisms enable GBMs to efficiently adsorb a wide range of antibiotics, making them highly effective in wastewater treatment applications.

GBMs are highly effective at removing antibiotics from wastewater because of their large surface area and versatile adsorption mechanisms. However, despite their efficiency, several limitations arise after their use, particularly concerning environmental and health risks. Key challenges include the regeneration of these materials for reuse, potential secondary contamination, and risks posed by the release of nanomaterials into the environment. Without proper management, these issues could undermine the environmental benefits of using GBMs in wastewater treatment.

To mitigate these risks, several strategies are being explored. Safe disposal methods, such as encapsulating GBMs to prevent antibiotic and nanomaterial leakage, are essential for preventing secondary pollution. Improved regeneration technologies, such as green chemical processes or thermal treatments, can extend the life of GBMs without compromising their adsorption capacity [122]. Research into biodegradable graphene composites also offers a future solution for reducing the long-term risks associated with nanomaterials. These approaches are critical for ensuring that GBMs remain a safe and sustainable option for wastewater treatment.

6. Conclusions

In conclusion, the increasing presence of antibiotics in wastewater presents significant environmental challenges, particularly in fostering the spread of antimicrobial resistance and impacting aquatic ecosystems. The advancements in graphene-based technologies from 2016–2024 have demonstrated substantial potential in addressing these issues, particularly through the utilization of GO, rGO, and various graphene composites. These materials have exhibited remarkable adsorption capacities and photocatalytic degradation efficiencies, particularly when functionalized or integrated with other substances such as metal oxides and polymers. Despite the challenges related to scalability, cost, and environmental impact, the progress in this field underscores the promising role of GBMs' ineffective antibiotic removal from wastewater. Future efforts should focus on optimizing these technologies for large-scale applications, ensuring their sustainability, and integrating them into broader environmental management practices. These advancements constitute critical steps toward safer and more sustainable approaches to managing wastewater contamination.

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