



Recent Advancements in Graphene-Based Nanocomposites for Enhanced Photocatalysis in Environmental Remediation: A Comprehensive Review

Jaspal Singh¹ · D. Duc Nguyen^{2,3} · Philippe Leclerc⁴ · Phuong Nguyen-Tri¹

Received: 24 September 2024 / Accepted: 12 May 2025

© The Author(s), under exclusive license to Springer Nature Switzerland AG 2025

Abstract

The advanced oxidation process provides significant advancements in the field of environmental remediation all around the world. In this context, various photocatalysts with different light-sensitive capabilities were engineered and employed for improved photocatalytic performance. Recently, the development of 2D-layered photocatalysts has attracted significant attention due to their tremendous pollutant removal efficiencies. The current review explicitly explains the recent developments in graphene and their derivatives-based heterojunction for enhanced photodecomposition behavior for water purification and detoxification. It will provide a systematic roadmap to develop advanced graphene-based nanocomposite for future technologies. In addition, the review highlights the different synthesis strategies, properties, charge transfer mechanisms, and optimized parameters for different graphene-based photocatalysts. The insight perspective and the future possibilities from the recent reports on graphene, graphene oxide (GO), and reduced graphene oxide (RGO) have been presented here, which provides the potential to enhance their photodecomposition efficiency for water purification further. A comprehensive view of the different graphene-based heterostructures such as plasmonic nanocomposites, semiconductor nanocomposites, transition metal dichalcogenides (TMDCs)-based nanocomposites, and organic molecules-based photocatalyst and their underlying charge transfer mechanism have been presented. Finally, a summary and future perspective of the development of graphene-based photocatalysts has been mentioned and elaborated. The advancement of photocatalytic systems, integration with graphene-based nanostructures, and innovative technologies for large-scale implementation are poised to offer solutions to energy and environmental challenges in the foreseeable future.

✉ D. Duc Nguyen
nguyensyduc@gmail.com

✉ Phuong Nguyen-Tri
Phuong.nguyen-tri@uqtr.ca

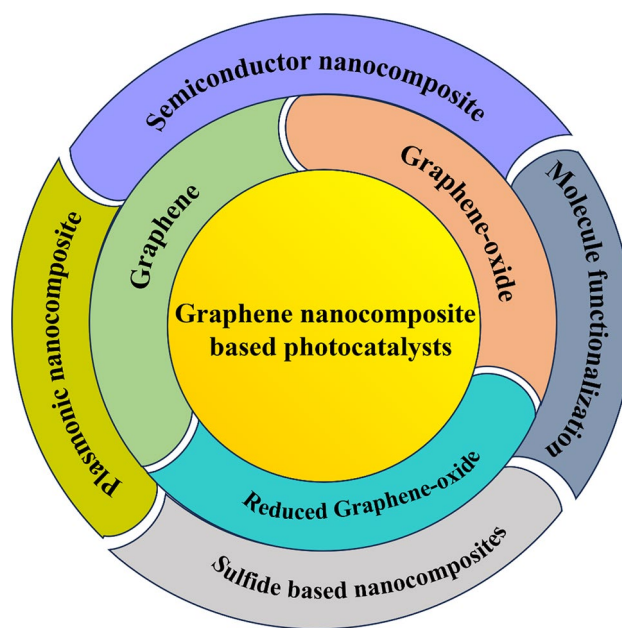
¹ Laboratory of Advanced Materials for Energy and Environment, Université du Québec À Trois-Rivières (UQTR), 3351, Boul. Des Forges, C.P. 500, Trois-Rivières, QC G9A 5H7, Canada

² Department of Civil & Energy System Engineering, Kyonggi University, Suwon-si, Republic of Korea

³ School of Engineering & Technology, Duy Tan University, Da Nang, Vietnam

⁴ Laboratory for Physics of Nanomaterials and Energy (LPNE), Research Institute in Materials Science and Engineering, University of Mons (UMONS), 7000 Mons, Belgium

Graphical Abstract



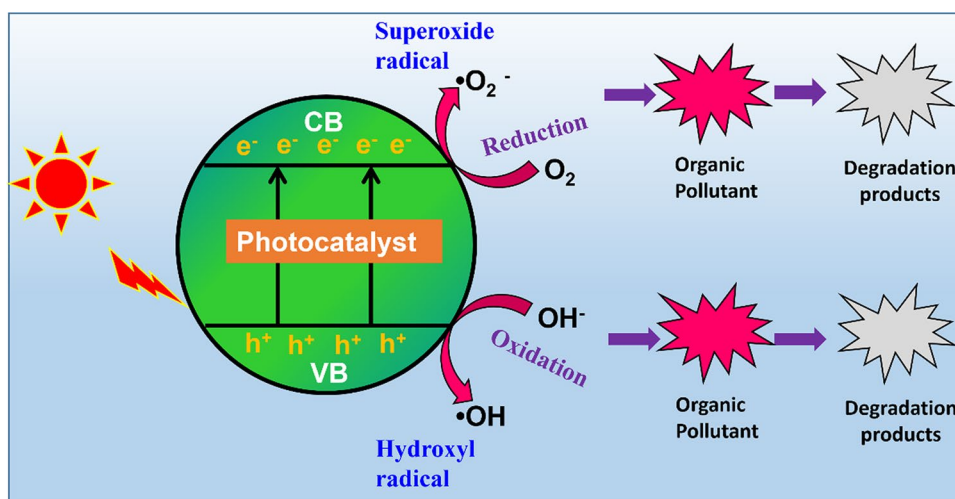
Keywords Graphene-based photocatalysts · Photocatalysis · Nanocomposites · Water remediation · Graphene-based photocatalysts

Introduction

All around the world, environmental remediation is one of the prime objectives for researchers due to the sustainability of societies and humankind. Due to the continuous growth in the population and industrialization, several water resources are either contaminated or ruined. Several pharmaceutical, chemical, and textile industries are responsible for water pollution. Several untreated hazardous long-chain organics, such as azo dye molecules, are carcinogenic and pollute water resources (Khin et al. 2012; Liu et al. 2022; Göktaş et al. 2014; Goktas et al. 2022a; Aslan et al. 2024). These highly stable organic molecules not only harm the living species in the water but are also responsible for dangerous diseases such as diarrhea, typhoid, and hepatitis A, which can drastically damage human health (Felis et al. 2022; Dridi et al. 2024). Apart from this, these highly stable organic molecules disturb the biochemical processes of the plants (Schweitzer and Noblet 2018). Consequently, the food chain systems of the environment are drastically affected and diminished. Different organic pollutants can be persistent in the water for a long time and are therefore responsible for soil pollution (Gao et al. 2022). Thus, to minimize and control human health risks, there is an urgent need for effective ways to remediate the water sources and help to maintain

the quality of drinking water; several agencies such as the World Health Organization (WHO), Environmental Protection Agency (EPA), and the European Union (EU) implemented some strict rules (Cousins et al. 2022; Dodds et al. 2020). They suggested using an advanced oxidation process to detoxify various highly stable organic molecules that persist in the water sources. In the advanced oxidation process (AOP) under light exposure, with the presence of the suitable photocatalyst material, the organic moieties become decomposed due to the redox reaction (Yang et al. 2022; Singh et al. 2023a). Advanced Oxidation Processes (AOPs), first used in the 1980 s to treat drinking water, have emerged as one of the most potent solutions to the contemporary pollution and energy crisis across the globe. AOPs are chemical reactions that generate and use reactive oxygen species (ROS), most commonly hydroxyl radicals, to target an analyte, usually a toxin, and convert it into a non-toxic form (Singh et al. 2021a, b, 2023a, c). Hydroxyl radicals are one of the most reactive oxidizing agents, with an oxidation potential between 2.8 and 1.95 V (pH 0–14) when measured against the saturated calomel electrode (Fujishima and Honda 1972; Singh and Soni 2020a, b, c). Their rapid, non-selective attack can disintegrate any analyte by forming other reactive species, such as H_2O_2 and/or superoxides ($\text{O}_2^{\cdot-}$). Since hydroxyl radicals are very short-lived, they are generated in situ during the reaction through oxidizing agents, irradiation, catalysts, and/

Fig. 1 Schematic representation of the photocatalytic process mediated by photocatalysts



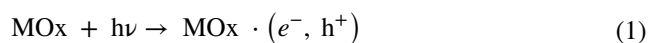
or their combinations—different AOPs, such as hydroxyl radical-based, ozone-based, Fenton-based, UV-based, are known, studied, and applied. Different types of ROS can be generated depending on the AOP process (Parrino et al. 2020; Singh et al. 2020c; Singh and Soni 2021c). For example, for the ozone treatment process, several reactive species such as $\cdot\text{OH}$, $\text{HO}_2\cdot$, $\text{HO}_3\cdot$, $\cdot\text{O}_2^-$ and $\cdot\text{O}_3^-$ were created, which interacted with the complex organic moieties and decomposed them into H_2O and CO_2 molecules (Kumar et al. 2022).

Primary deciding factors for process choice depend on the concentration of oxidizing agents, pollutants, the catalyst used, efficiency, irradiation type and intensity, wastewater quality, and most importantly, process cost, which includes operation, production, and management expenditures.

As an efficient and low-cost solution, nanomaterial-mediated photocatalytic AOP became popular for remediating polluted water bodies, which mainly constitute organic pollutants and textile discharges. AOP requires no heavy machinery and multi-step detoxification like conventional methods. In addition, the advanced oxidation process requires only a suitable light source, and the Sun serves as a readily available light source that provides highly intense light with different ranges, mainly from UV, visible, and NIR. Depending upon the optical absorbance ability of the photocatalyst, incident light of a particular wavelength can be used. Since sunlight is primarily visible and NIR light, the photocatalyst materials sensitive to these ranges attracted great interest. Additionally, various materials have been employed to harvest most of the sunlight illumination.

Nanoscale photocatalysts, mainly metal oxides, have a high surface area to volume ratio, high degradation efficiency, robust UV sensitization, good chemical stability, easy synthesis, environmental benignity, and cost-effectiveness, which explains their selection over other AOP alternatives. Titanium dioxide (TiO_2) was the first and the most

common photocatalyst used for treating contaminated water owing to its excellent charge carrier separation efficiency. However, many other suitable alternative oxide materials, such as zinc, indium, copper, molybdenum, have been developed into functional photoactive catalysts (Chan et al. 2011; Singh et al. 2020d; Singh and Soni 2021d, Singh et al. 2021e, f; Zhang et al. 2023; Göktaş et al. 2023). Although majorly designed for treating chemical contaminants, the photocatalytic ability of such photocatalysts is also being used for the inactivation of bacterial contaminants, air purification, hydrogen production, sensing, fabricating re-useable surface-enhanced Raman spectroscopy (SERS) substrates, among others (Singh et al. 2019a; Singh et al. 2019b; Naldoni et al. 2018; Singh et al. 2021f; Yan et al. 2023; Wang et al. 2024a, b; Liu et al. 2024; Ji et al. 2016; Pogacean et al. 2015; Göktaş et al. 2023; Sahin et al. 2024). Principally, all semiconductor photocatalysts are photoexcited with a light source, generating ROS, which reacts with toxins adsorbed on the catalyst surface, leading to the degradation of the toxin (Göktaş et al. 2023; Sahin et al. 2024; Singh et al. 2023c) following the pathway as shown schematically in Fig. 1. Mathematically, this can be represented as:



Although metal oxide systems are sought as the most powerful photocatalytic sources, they are often used hesitantly due to their fast charge carrier recombination and limited light sensitivity. However, by any means, if the generation, separation, and delocalization of the charge carriers can be enhanced, the production of reactive species and, hence, the degradation efficiency of the catalyst can be increased. The need for visible light sensitivity arises when these photocatalytic systems are to be used under natural light sources such as sunlight, which is mainly made up of visible

spectrum; under artificial light sources, only delayed recombination would work fine to achieve higher degradation.

To overcome the limited effective surface area of the metal oxide, 2D-layered nanostructures have come into the limelight. Due to 2D-layered structures and effective enhanced surface area ($2600 \text{ m}^2 \text{ g}^{-1}$), they interact more effectively with organic moieties (Ravi et al. 2020; Garg and Chandra 2022). Graphene is pure carbon-based nanostructure with honeycomb structures, which show a semi-metallic nature due to zero bandgap (Han et al. 2020; Ibrahim et al. 2021). The existence of π -state valence band (VB) and π^* -state conduction band (CB) and their convergence at Dirac point (or Brillouin zone) enables the graphene to exhibit outstanding charge mobility and exceptional conductivity (Zhang et al. 2015). In addition, this novel band structure of graphene offers an electron migration speed of $200,000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (Garg and Chandra 2022). Moreover, the graphene layer exhibits excellent thermal conductivity and mechanical strength, indicating good stability (Singh et al. 2023 d). The atomic layer 2D thickness of graphene offers improved charge separation owing to their efficient interaction with incident light (Li et al. 2023). The graphene layer can capture photoinduced electrons and act like an electron acceptor during the photocatalysis process. Consequently, it can lower the charge recombination process, highlighting its advantage over the conventional photocatalyst (Han et al. 2016; Yang et al. 2014, 2023).

Due to the above-mentioned fascinating properties of graphene layers, their attachment to the photocatalyst semiconductor, such as TiO_2 , ZnO , CuO , and MoS_2 , provides excellent sensitization and rapid photoinduced electron transfer; consequently, the recombination rate of the photocatalyst is quenched (Yang et al. 2023; Lu et al. 2016). Moreover, graphene attachment boosted the effective surface area, facilitating interaction among the organic moieties and photocatalysts. Thus, integrating graphene with different semiconductors can significantly attain an effective surface area and efficient charge separation, significantly enhancing photocatalytic activity. Different graphene-based nanohybrid systems have been developed for enhanced artificial photosynthesis processes. These unique systems improved performance considerably for different photocatalyst processes such as water detoxification, conversion of selective organic molecules, CO_2 reduction, and water splitting (Yang et al. 2023; Lu et al. 2018; Khan et al. 2022; Quan et al. 2017).

Since its discovery till date, several routes have been designed to achieve effective charge separation for better photocatalytic efficacy, of which one has been attaching graphene (G) and its related moieties, such as Graphene Oxide (GO) and Reduced Graphene Oxide (RGO), to metal oxide photocatalysts (Padmanabhan et al. 2021). G, GO, or RGO has been associated with reduced bandgaps in metal oxide, improved light sensitivity, excellent electron

scavenging activity, increased direct charge transfer, and thus delayed charge recombination tendencies. The detailed description of Graphene, its properties, and its mechanism for improved photocatalysis is explained in the following sections of the review, along with examples to bring about better understanding to the reader both conceptually and visually.

Successful exfoliation of a layer of carbon atoms packed into a 2D honeycomb crystal lattice, better recognized as graphene today, has triggered excitement across scientific and technological societies since its discovery in 2004 (Fang et al. 2020). Considering its novel and intriguing properties, which include improved electrical conductivity, superior mobility, exceptional optical transmittance, higher chemical stability, and specific surface area (*theoretically*), its acceptance in various fields, including photocatalysis, is quite justified (Kuang et al. 2020). The introduction of graphene into a semiconductor matrix improves the catalytic performance of the composite by harnessing a co-catalyst role, which allows effective charge separation and transfer in semiconductors (Zhang et al. 2015).

Pristine graphene synthesis has been realized by epitaxial growth (Song et al. 2022), plasma etching (Li et al. 2022a, b), chemical vapor deposition (Shi et al. 2022), arc discharge (Shafiee et al. 2022), etc.,. However, for cases where graphene is utilized as an adjunct, such as in the case of photocatalysis, its processability with other compounds becomes vital given the final composite characteristics and properties. Notably, since most graphene-based photocatalyst fabrication is done in the liquid phase, solution-based techniques for graphene synthesis, mainly graphene oxide reduction to form GO or RGO, are standard. Oxygenated functional groups in GO allow flexible and easily accessible wet chemistry processability (Yang and Xu 2013b; Tu et al. 2013; Xiang and Yu 2013; Zhang et al. 2020a, b, c, d). Although, at present, there are no conventional criteria to categorize graphene-based compounds, depending on the method utilized to derive graphene, we can categorize it as:

- Reduced graphene oxide (RGO)—graphene produced by graphene oxide reduction,
- Solution exfoliated graphene (SEG)—graphene produced by graphite liquid exfoliation in suitable solvents,
- Organically synthesized graphene (OSG)—graphene obtained by organic synthesis.

Abundant chemical modification opportunities in graphene due to its flexible surface characteristics allow a variety of methods that could be used to fabricate graphene-based nanocomposite photocatalysts. Hydrothermal, solvothermal, sonication-assisted, and electrochemical deposition are a few to mention (Sui et al. 2020; Bhushan et al. 2020; Yan et al. 2021; Chen et al. 2020a).

All these methods have been exploited to twin graphene as a 2D support to form composite photocatalysts, which can be done in two different ways: *in situ* and *ex situ* (Zhang et al. 2015; Yang et al. 2014; Mohaghegh et al. 2015). During *in situ* synthesis, soluble precursor salts of the photoactive materials, GO, and other organic chemicals are mixed, followed by thermal, optical, chemical, or ultrasonic treatment to form nanostructure functionalized graphene composites. Specific synthetic conditions can be tuned to modify the size and shape of the photoactive component to form particles, wires, plates, or anisotropic. *In situ* methods generally do not require additional surfactants or linker molecules to form the composites, leading to direct interfacial contact between the complex's photoactive and the graphene components. The process overall is cheap, easy, and cleaner. As in the previous case, *ex situ* methods employ GO precursors and pre-synthesized or often commercially available photoactive components rather than their precursor salts. Since the photoactive components are performed to form a strong interfacial bond with the graphene component, this method generally adopts pre or post-treatment steps such as surface charge modification, chemical or thermal treatment, rendering the *ex situ* method less efficient. However, since the photoactive material is preformed, *ex situ* methods give better size and morphology control. The structure size and shape can be maintained uniformly throughout the process, allowing the required reproducibility of the procedure and the performance of the devised nanocomposite systems.

Although different graphene nanocomposite-based excellent reviews have been reported by several research groups (Ji et al. 2016; Goktas and Goktas 2021; Han et al. 2020; Zhang et al. 2015; Yang et al. 2014; Lu et al. 2016; Padmanabhan et al. 2021), which emphasize the preparation methods, functionalization, and diverse photocatalytic applications. This review mainly focuses on the surface engineering of graphene-based nanocomposites and offers a clear roadmap to obtain enhanced photodecomposition performance for water detoxification applications. The current review highlights the explicit exploration of the critical properties of graphene and its nanohybrids, which strongly influence its photocatalytic performance. In this review, recent progress in integrating graphene and its derivatives with other nanostructures such as semiconductors, noble metal nanoparticles, TMDCs, and organic molecules for boosted photodecomposition has been elaborated in detail. This review provides an integral overview that provides a clear understanding of the changes in the fascinating properties of graphene with surface modification by different nanostructures and their co-relationship with photodecomposition performance. We discuss several strategies for designing graphene-based nanohybrids and compare their photodecomposition efficiency, which provides a clear understanding for the readers and will help

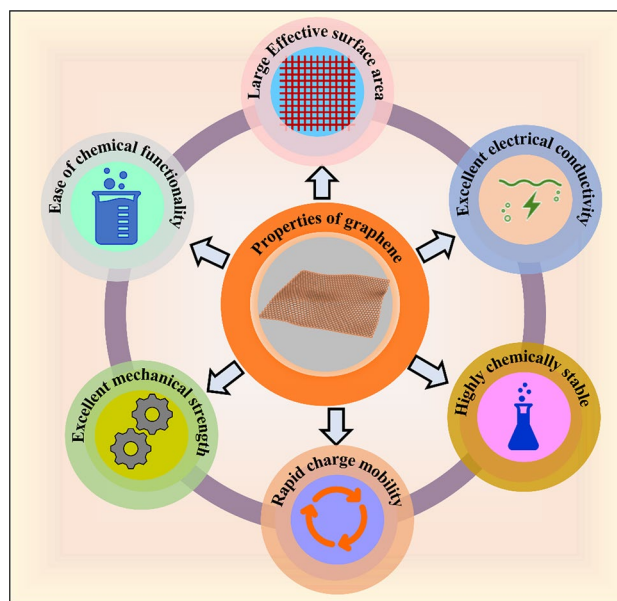


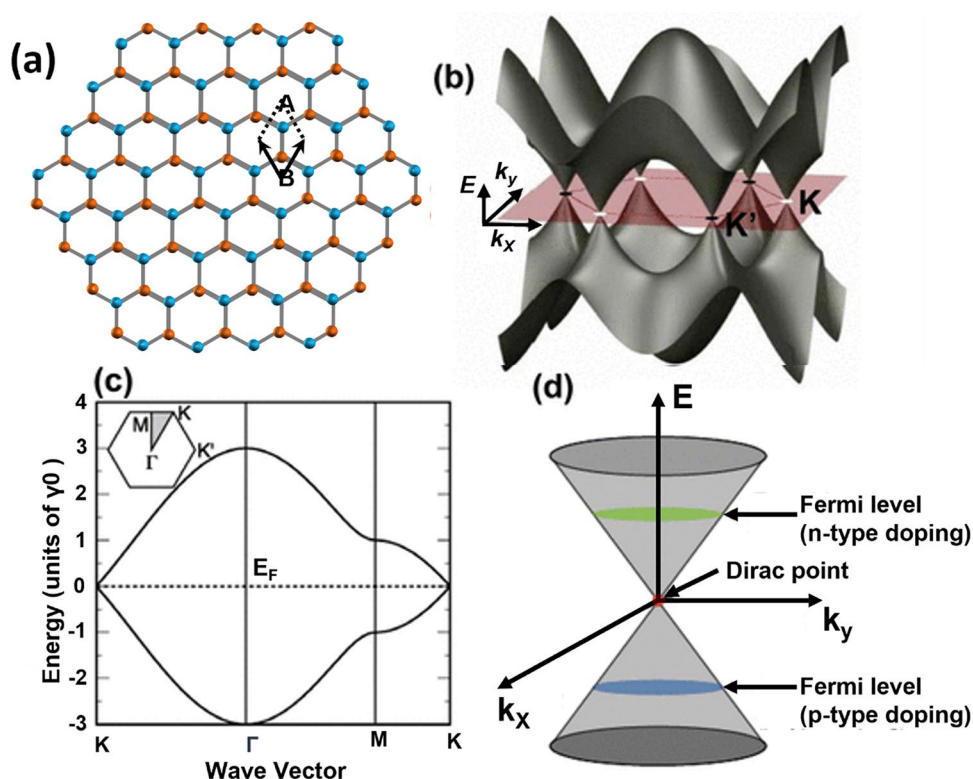
Fig. 2 Schematic diagram illustrating the key properties of graphene, which are essential for the photocatalytic process

develop graphene-based advanced photocatalysts further. Finally, we present a brief outlook on future challenges and potential advancements in the rising field of graphene-based photocatalysts.

Properties of Graphene and Their Derivatives

The fundamental properties of graphene and its derivatives, such as work function, exceptional electrical conductivities, strong electron-accepting capabilities, high transparency, and surface chemical and physical characteristics, depend on their novel surface, structural, and interfacial profiles. These properties include electronic structures, molecular adsorption, surface chemistry, and atomic arrangements, which influence the photocatalytic efficiencies of the graphene and graphene-based derivatives. Interestingly, the chemical modification on the surface of graphene gives rise to significant tunability in their electrical properties and optical properties (Li et al. 2016) consequently, photodecomposition activity can be improved. The overview of outstanding properties of graphene, which make it beneficial for the photocatalysis process, has been depicted in Fig. 2. Apart from this, the various other properties that highly enhance the photodecomposition performance of graphene and their derivatives, such as GO and RGO will be discussed in detail in the current section.

Fig. 3 **a** Honeycomb lattice structure of graphene with two atoms, namely A and B per unit cell, **b** 3D view of the band structure of graphene, **c** dispersion of the band structures of graphene, **d** low energy band structures of graphene indicating the reverse cone touching with each other at Dirac point. They reproduced with permission (Avouris 2010) Copyright 2010 American Chemical Society

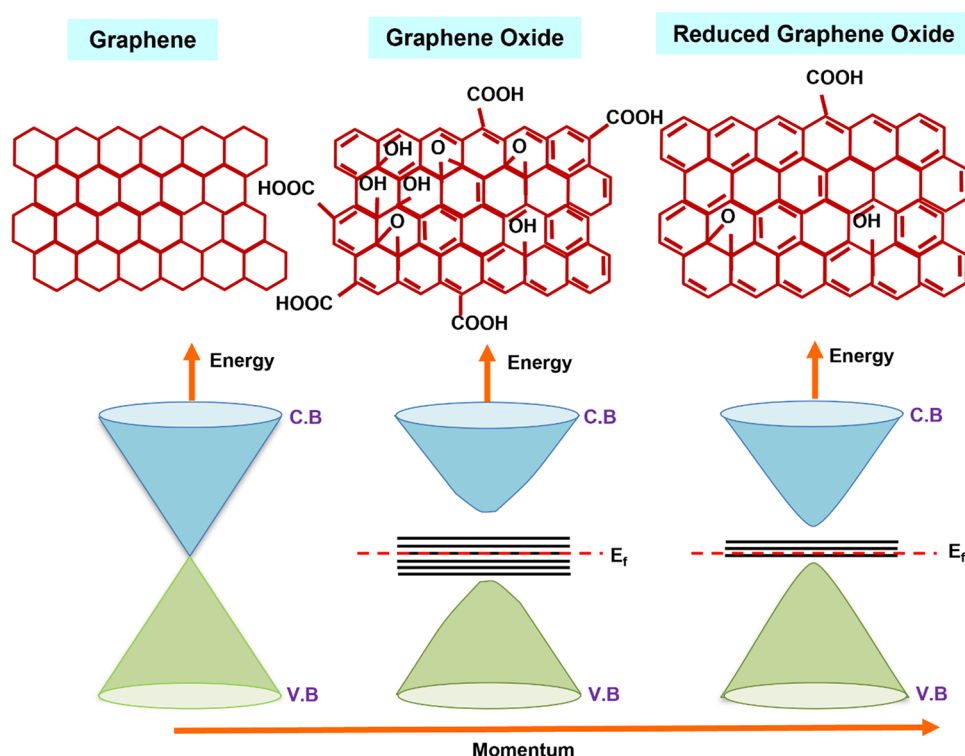


Crystallographic Properties

The atomic structure properties of graphene have majorly contributed to enhancing photocatalytic behavior. This section presents the key crystallographic properties of graphene for improved photocatalytic activity in depth. The graphene contained the 2D-layered structure, which gave rise to its effective surface area (theoretically calculated $\sim 2630 \text{ m}^2/\text{g}$), which provided a high number of active sites and facilitated different organic moieties to attach to the surface and enhance the photocatalytic activity (Zhang et al. 2020a, b, c, d). Additionally, 2D-layered graphene affirms the high electron mobility ($20,000 \text{ cm}^2/\text{V}\cdot\text{s}$) and can significantly improve photocatalytic activity through efficient charge migration (Garg et al. 2022). Due to sp^2 hybridization in graphene, the carbon atoms form in-plane σ bonds while the rest of the p orbitals create the π bonds, giving rise to graphene's unique electronic structures (Ji et al. 2016; Garg and Chandra 2022). In addition, the extensive employment of graphene in the photocatalytic processes is due to their fascinating electronic properties, driven by the 2D-layered crystal lattice of graphene. For the structure, the valence orbitals of the carbon atoms exhibit the sp^2 hybridization. Due to this hybridization, three planar sigma orbitals are evenly spaced at 120° angle with each other. Apart from this, another single 2p orbital is directed toward the perpendicular of the plane of a graphene sheet (Li et al. 2022a, b). Thus, this unique atomic arrangement

generates the honeycomb-like structure of graphene, which provides a suitable platform to attach with the other different nanostructures, such as nanorods, quantum dots, 2D-layered nanostructures, and 3D-nanostructures (Lee et al. 2012; Xiao et al. 2014; Vazirisereshk et al. 2019; Xiong et al. 2021). Lee et al. (2012) employed the sol-gel method and explored the attachment of TiO_2 nanorods over the graphene sheets. Their study highlighted that the high surface area of graphene increased the probability of the TiO_2 nanorods attachment. In another report, Xiao et al. (2014) demonstrated the formation of CdS quantum dots functionalized graphene sheets. They showed the layer-by-layer assembly of CdS QDs on the poly(allylamine) functionalized graphene layer. The CdS/graphene nanohybrids exhibited improved photocatalytic and photo-electrochemical properties. The hexagonal honeycomb lattice of graphene, with two carbon atoms per unit cell, results in a unique band structure first calculated by Wallace in 1947 (Wallace 1947) (Fig. 3a, b). From an ideal 2D system, the π -electrons in graphene are explored by a single atom thickness and noninteracting π and π^* states. The π -states generate the valence band, while the conduction band is produced by the π^* states in graphene. These two bands touch at six Dirac or neutrality points, which can be reduced to a pair, K and K', independent of each other (Fig. 3c). The bands exhibit a linear behavior at low energies, which is most relevant in electron transport. In the context of the electronic properties of graphene, the zero bandgap semiconductor behavior is explained based on

Fig. 4 Pictorial view showing the variation in the lattice of graphene, GO, and RGO with the changes in their band structures. They reproduced with permission (Abid et al. 2018) Copyright 2018 Springer Nature



the contact of the two bands at Dirac points (Pramanik et al. 2022; Pham et al. 2022) (Fig. 3d).

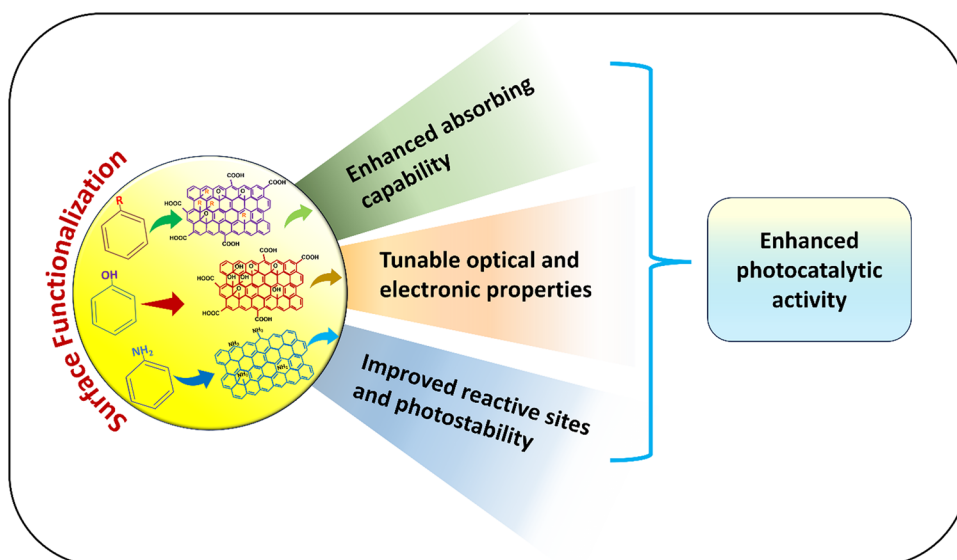
In graphene structures, C–C bonding provides outstanding strength and increases its mechanical properties (Young's modulus ~ 1 TPa). This mechanical strength allows the 2D-layered graphene structure to generate robust support in different photocatalytic structures (Garg et al. 2022; Papa-georgiou et al. 2017). Busarello et al. (2023) recently fabricated graphene-supported ZnO quantum dots (QDs) using the wet chemical method. They turned the energy harvesting properties of these nanohybrids by varying the size of ZnO QDs over the surface of graphene. They determined that ZnO QDs supported by graphene sheets with optimum size (4.5 nm) enhance photocatalytic activity compared to pristine ZnO QDs.

Optical Properties

The 2D-single layer of graphene can absorb 2.3% of the visible region, allowing it to graphene in visible light transparently. This fascinating optical behavior of graphene permits visible light to pass through it and interact with the photocatalysts (for example, ZnO, TiO_2 , CuO, and CdS) wrapped or underneath graphene (Lu et al. 2021; Wang et al. 2017). Apart from this, it has been reported that pure graphene can harvest the UV region due to zero bandgap, but with the incorporation of defect states or functionalization with different groups ($-\text{OH}$, $-\text{COOH}$, and pyridine), their absorption edge can be

extended up to the visible region. It can boost the photocatalytic process (Georgakilas et al. 2012). Recently, Dubey et al. (2024) reported the fabrication of nitrogen-enriched graphene through the combination of thermal annealing and wet chemical methods. Their study demonstrated the improved optical absorption and efficient charge separation of graphene due to functionalization with the nitrogen group. The modified graphene exhibited outstanding photocatalytic activity by degrading orange-G under UV light. In another study, Li et al. (2023) showed the preparation of TiO_2 /poly(diallyldimethylammonium chloride)/graphene quantum dot hybrid through the hydrothermal method with the chemical method. Their study highlights the improvement in the photocatalytic performance of these nanohybrids when graphene quantum dots are functionalized with a hydroxyl group. They have also highlighted that the functionalized group enhanced the number of active sites and improved the interfacial charge separation in TiO_2 /PDDA/QDs photocatalyst, which is majorly responsible for enhanced photocatalytic activity. The optical profiles of graphene, graphene oxide, and RGO are fascinating due to their applications in various fields. Graphene contains a single layer of carbon atoms and exhibits unique optical transitions due to its zero bandgap (Avouris 2010). On the other hand, graphene oxide consists of oxygen functional groups, which are semiconductors, resulting in the broader bandgap observed (Fig. 4) (Abid et al. 2018). The bandgap of graphene oxide is estimated at 1.6–2.5 eV

Fig. 5 Pictorial view indicates the improvement in the different properties of graphene for enhanced photocatalytic activity



(Velasco-Soto et al. 2015; Moon et al. 2010; Guo et al. 2022). Velasco-Soto et al. (2015) studied the variations in bandgap due to changes in eco-friendly reagents such as glucose, fructose, ammonium hydroxide, and ascorbic acid. They have shown that glucose and fructose show reactivity under the presence of ammonium hydroxide, while ascorbic acid can reduce GO independently. Their study demonstrated the systematic procedure to tune the bandgap of the graphene oxide by modification using the chemical route. They concluded that at a pH value of 10, the bandgap of graphene oxide varied from 2.7 to 1.15 eV. Reduced graphene oxide, obtained from graphene oxide, also possesses unique optical properties due to the restoration of the graphene structure. The optical absorption peak is generated for RGO due to $\pi \rightarrow \pi^*$ transition in the carbon ring (Lu et al. 2018). The bandgap of reduced graphene oxide is narrower than GO and varies from 1.1 to 1.4 eV (Lu et al. 2018). Abid et al. (2018) synthesized the RGO from Feng et al. 2013GO and studied the variations in their bandgap. For RGO samples, they observed the bandgap varied from 1 to 1.69 eV, which is controlled by the content of the oxygen atoms. They have concluded that the bandgap is adjusted depending on the oxygen atoms in RGO. Nugraheni et al. (2015) observed a reduced energy gap of RGO by changing the oxygen content. They prepared the RGO using the wet chemical method and studied the effect of oxygen content variation by the thermal annealing process. By varying the temperature from 400 °C to 600 °C, the energy gap of RGO changes from 0.14 to 0.0.67 eV. The bandgap of RGO value is strongly influenced by the degree of the reduction. During the reduction, the oxygen atoms' density varies, affecting the bandgap and electrical conductivity

of the RGO (Feng et al. 2013; Nugraheni et al. 2015; Yu et al. 2020).

Surface Chemical Properties

2D-layered graphene derivatives contained different defect states and functional groups such as epoxy, hydroxyl, carboxyl, and amino groups (Wang et al. 2018a, b). These existing functional groups enable the adsorption ability of graphene oxide and RGO by chemical interaction or physical adsorption ($\pi \rightarrow \pi$ stacking interaction). It has been explored that functional groups exist over the surface not only to remove the long chain containing azo dye molecules but also to be capable of adsorbing heavy metals, non-metal ions, and gaseous state pollutants (Li et al. 2016; Nemati et al. 2022). Figure 5 shows the enhancement in the absorbing ability of graphene owing to the increment in the reactive sites and improvement in the optical properties, which significantly improves their photocatalytic activity. It represents that the presence of the alkyl group (R) can enhance the absorbing capability of graphene while the presence of OH and NH₂ groups over the surface of graphene can improve the optical properties and photostability of graphene, respectively (Anichini and Samorì, 2021; Shah et al. 2024; Borane et al. 2024).

Guirguis et al. (2020) used the microwave plasma chemical vapor deposition method to fabricate a defect-rich graphene layer. They used thermal annealing at 800 °C to improve the crystallinity. They have pointed out that enhanced photocatalytic activity can be attributed to the efficient charge transformation among oxygen functional groups and dangling bonds appearing around the defective sites. In their photodecomposition test, a 5-ppm solution of methylene blue (MB) pollutant was decomposed under

UV light illumination using defect-rich graphene substrates with a size of 2 cm × 2 cm. The origin of the outstanding adsorption ability of graphene-based derivatives is due to an effective high surface area, which contains many active sites all over the surface.

Moreover, enhanced active sites on the surface of graphene derivatives significantly increase the interaction with pollutant molecules (Lyu et al. 2023). Therefore, the photoinduced electron–hole pair and unsaturated radicals (hydroxyl and superoxide) efficiently interacted with the organic molecules and decomposed them rapidly. Surface functionalization of graphene with different groups provides tunable optical properties and increases structural durability under light illumination. Pristine graphene structure tends to damage or decompose under UV light exposure. With the attachment of a functional group or some dopant, the rigid framework helps to protect the graphene structure from oxidation and decomposition under intense light illumination (Zhang et al. 2015; Abid et al. 2018).

On the other hand, doping with different nonmetals, such as nitrogen and sulfur, in to graphene can tremendously improve the charge mobility and light-harvesting ability (Jiang et al. 2019). It has been reported that N-doped graphene is suitable for CO₂ capture and H₂ production applications due to its outstanding adsorption ability toward the H⁺ ions and CO₂ molecules (Lyu et al. 2023; Jiang et al. 2019; Liu et al. 2023; Kemp et al. 2013). Kemp et al. (2013) reported synthesizing N-functionalized graphene from activating RGO/polyaniline nanocomposite. The effective surface area of the N-graphene is 1336 m²/g and absorbs 2.7 mmol/g of CO₂ gas. They have optimized the N doping for enhanced adsorption capability. The prepared optimized sample also shows the sensitivity toward the other gases such as N₂, H₂, Ar, and CH₄.

Semiconductor Properties

To employ graphene for several semiconductor-based applications, different research groups have explored its semiconductor properties (Lu et al. 2013; Radadiya 2015; Xiang et al. 2012). As mentioned in Sect. "Optical Properties", the functionalization of graphene with other functional groups provides control of the electrical properties and variations in the bandgap, which majorly affects the photocatalytic profiles. Different ways, such as multilayer graphene formation, surface functionalization, confinement, and molecular adsorption, have revealed the semiconductor nature of graphene (Xiang et al. 2012; Di et al. 2016). Different functional groups have been demonstrated to make graphene, either n-type or p-type semiconductors (Li et al. 2016; Xiang et al. 2012; Di et al. 2016; Sreepasad and Berry 2013). The attachment of the oxygen atom over the surface of graphene breaks their sp² conjugate network and restricts the original

π-electrons within the separated sp² domains (Nebol'Sin et al. 2020). On the other hand, the π* antibonding maintained its position as previously. Moreover, the position of the conduction band minima of graphene oxide is unaffected, while the valence band maxima shifted due to the interaction between C and O atoms (Hsiao et al. 2010). Based upon the surface functionalization of graphene with oxygen atom content, tunability has been achieved in the bandgap for the case of reduced graphene oxide. As mentioned above, graphene oxide with varied CB and VB positions and semiconductor nature actively provides outstanding performance in different photocatalysis processes such as water splitting, CO₂ capturing, and H₂ production (Xiang et al. 2012; Pei et al. 2020). Thus, graphene-based derivatives offer the flexibility to tune the CB and VB positions according to the application. Similarly, B doping in graphene also provides improved charge transformation and is beneficial to improve the photodecomposition process of GO (Singh et al. 2018). Singh et al. (2018) reported the formation of B-doped graphene using the modified Hummer's method. Their study mentioned that improved photodecomposition activity could be assigned to the boosted density of charge carriers in the valence band due to the p-type nature of graphene oxide. In their photocatalytic studies, methyl orange and methylene blue were considered as the pollutant solution. They used 25 mg of photocatalyst sample to decompose 10 ppm of methyl orange (MO) and MB under UV light illumination for 100 and 50 min, respectively. In another report, Tai et al. (2024) used the photoreduction method to fabricate nitrogen-doped photo-reduced graphene oxide and varied the nitrogen doping content for improved photocatalytic activity. They also concluded that the enhanced photocatalytic activity originated due to high free-electron carrier density and a solid n-type conductivity. Luo et al. (2018) synthesized sulfur-doped graphene QDs/TiO₂ nanohybrids using the facile hydrothermal method and employed them to decompose MO pollutants under UV light illumination. In their photocatalytic test, 20 mg of S-doped QDs/TiO₂ photocatalyst was decolorized in a 20 ppm solution of MO for 8 min. They have highlighted that improved optical absorption and efficient synergistic effect are majorly responsible for boosting photocatalytic performance.

Semi-metallic Properties

Graphene 2D layers show a semi-metallic nature, which makes them unique from typical metals and semiconductors. As discussed in Section "Crystallographic Properties", the VB and CB touch each other at the Dirac point, making the graphene a zero bandgap semiconductor. In the graphene structure, near the Dirac points, the electron's energy is linearly proportional to the momentum (Gallerati 2022). The charge carriers near the Dirac point act like massless

fermions and attain high speed ($\sim 10^6$ m/s). Apart from these fascinating properties, the charge carriers can move speedily up to long distances without showing scattering, indicating graphene's ballistic transport properties (Ji et al. 2016; Garg and Chandra 2022). These outstanding charge migration behaviors and fascinating conductivity profiles of graphene make them suitable for photocatalysis. Graphene exhibits a semi-metallic nature due to its high tendency to accept electrons, which improves the photodecomposition ability of photocatalysts. In graphene-semiconductor heterostructures, graphene facilitates the electron transfer from the semiconductor to graphene through the proper band alignment (Minella et al. 2017; Liu et al. 2017). Zhang et al. (2020a, b, c, d) demonstrated the enhanced photodecomposition activity of Cu_2O /graphene oxide nanohybrids prepared by the green precipitation method. Their study highlighted the 1.6 times better photocatalytic activity of Cu_2O /graphene than that of pristine Cu_2O . The boosted decomposition activity is assigned to the efficient charge transformation from Cu_2O to graphene oxide. The band alignment depends upon the work function of the materials and can be tuned by varying the material's work function. The work function is a crucial parameter defined as the minimum energy needed to eject the electron (Song et al. 2012). For the development of graphene-based photocatalysts, the variation in graphene offers outstanding control over the charge transfer mechanism. The reported work function of graphene is similar to graphite ~ 4.42 eV, which can be tuned by doping certain elements (Li et al. 2016; Song et al. 2012). For example, the upshift was achieved by doping some metal chloride, while nitrogen doping in graphene reduces their work function (Kwon et al. 2012; Rybin et al. 2016). Thus, the work function tuning provides sufficient control to change the band alignment of graphene-based photocatalysts. Recently, Jiang et al. (2023) used the hydrothermal method to fabricate CdS -RGO nanohybrids, which are further functionalized with anthraquinone-2-sulfonate, to improve photocatalytic activity further. They mentioned that boosted photocatalytic activity is ascribed to the interfacial charge transfer effect among CdS and RGO. Moreover, RGO also provided an increased number of active sites, which is also beneficial for their photocatalytic activity.

Factors Affecting the Photocatalytic Activity of Graphene

Apart from the properties mentioned above, several parameters, such as defect states in graphene, lattice strain, grain size, and annealing, are essential parameters that strongly influence their photodecomposition activity (Han et al. 2020; Zhang et al. 2015; Quan et al. 2017; Padmanabhan et al. 2021). For graphene sheets, the defect states in graphene can modify the electronic properties and trap the photogenerated

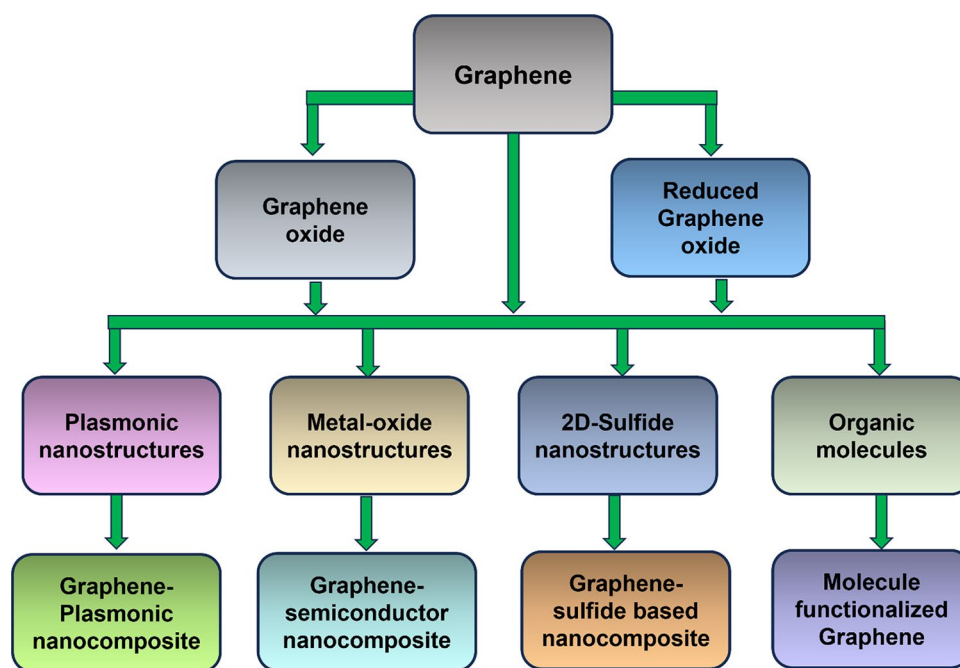
electrons, giving rise to efficient charge separation (Han et al. 2020). Also, due to the existence of defect states in graphene creates new active sites and boosts the adsorption activity of graphene (Ibrahim et al. 2021). The conductivity and electronic properties suffer a lot after the increment of defect states beyond the optimum density, resulting in decreased photodecomposition activity (Maarisetty and Baral 2020). Recently, Wang et al. (2024a, b) reported the role of defects in the improved activity of 3D-graphene-based composites. In their study, they attached $\text{TiO}_2\text{-C}_3\text{N}_4$ nanohybrids with 3D graphene by hydrothermal method and incorporated different defect states in TiO_2 , C_3N_4 and 3D graphene through plasma treatment. The obtained defective nanohybrid was employed for the decomposition of tetracycline. Their study highlighted that the defect creation in the $\text{TiO}_2\text{-C}_3\text{N}_4$ nanohybrids-3D-graphene increased the number of active sites and provided efficient charge transfer by capturing the light-driven charge carriers.

Grain size optimization is another critical aspect of the development of graphene-based photocatalysts. It has been reported that optimum grain size can further enhance the effective surface area of graphene, improving the active sites to interact with several organic molecules. Song et al. (2019a, b) prepared the $\text{TiO}_2/\text{P3HT}$ /graphene-based thin films by varying the grain size of graphene. They concluded that the optimum grain size ($150 \text{ m}^2/\text{g}$) showed the maximum photodecomposition activity for the RhB molecule solution. They have also highlighted that the optimum grain size of graphene boosted the effective surface area, resulting in photodegradation activity.

One of the other vital parameters is essential, like grain size and defect states. Thermal annealing temperature and atmosphere are also important factors influencing the photodecomposition activity of graphene-based nanohybrids (Huang et al. 2021). The annealing process in graphene reduced the microstrain and improved the crystallinity, improving its electrical conductivity significantly. Secondly, the annealing atmosphere provides the engineering in the defect states of graphene to interact with the different pollutant molecules. Sreeprasad and Berry (2013) prepared the SnS /graphene nanohybrids using a modified hydrothermal process and annealed the sample at 300°C in a hydrogen atmosphere and air. During the thermal annealing process, the defect concentration increased, which improved the number of active sites. Also, they observed that the photocatalytic activity improved for the SnS /graphene sample, which was annealed in a hydrogen atmosphere due to an efficient synergistic effect between SnS and graphene. For the SnS /graphene sample, annealed in the air indicates lower photocatalytic activity due to an increment in the oxygen defect states.

As mentioned above, graphene layers exhibit exceptional mechanical, electronic, and enhanced surface area

Fig. 6 Roadmap of graphene and its derivatives, showcasing various nanostructures designed to enhance photocatalytic activity



properties. Graphene, with its remarkable properties, enables efficient charge separation and can combine effectively with other nanostructures (Goktas and Goktas 2021). Additionally, the presence of graphene significantly increases the density of reactive sites. However, it has been observed that graphene's properties are not always superior when compared to other 2D-layered nanostructures, such as transition metal dichalcogenides (TMDCs), or one-dimensional nanostructures like carbon nanotubes (CNTs).

When comparing the bandgap properties of graphene and TMDCs, it can be noted that TMDC structures exhibit a bandgap of approximately ~ 1.7 eV with semiconducting nature, while graphene has a zero bandgap (Li et al. 2016; Manzeli et al. 2017). Furthermore, TMDCs display optical absorption spanning the visible to near-infrared (NIR) region, whereas graphene exhibits absorption primarily in the UV region. The electron mobility of graphene is significantly higher than that of TMDCs, implying that TMDCs have lower charge transfer efficiency compared to graphene. However, TMDCs excel in light harvesting and their inherent semiconducting nature makes them directly applicable in photocatalysis without the need for modification. In contrast, graphene often requires modifications to enhance its photocatalytic activity.

CNTs represent another type of interesting nanostructure with a chemical composition similar to graphene but with a one-dimensional (1D) morphology. This 1D structure allows CNTs to exhibit directional charge transport, which is absent in graphene. Studies have reported that multi-walled CNTs demonstrate enhanced mechanical and thermal stability compared to graphene (Wu et al. 2020). However, the

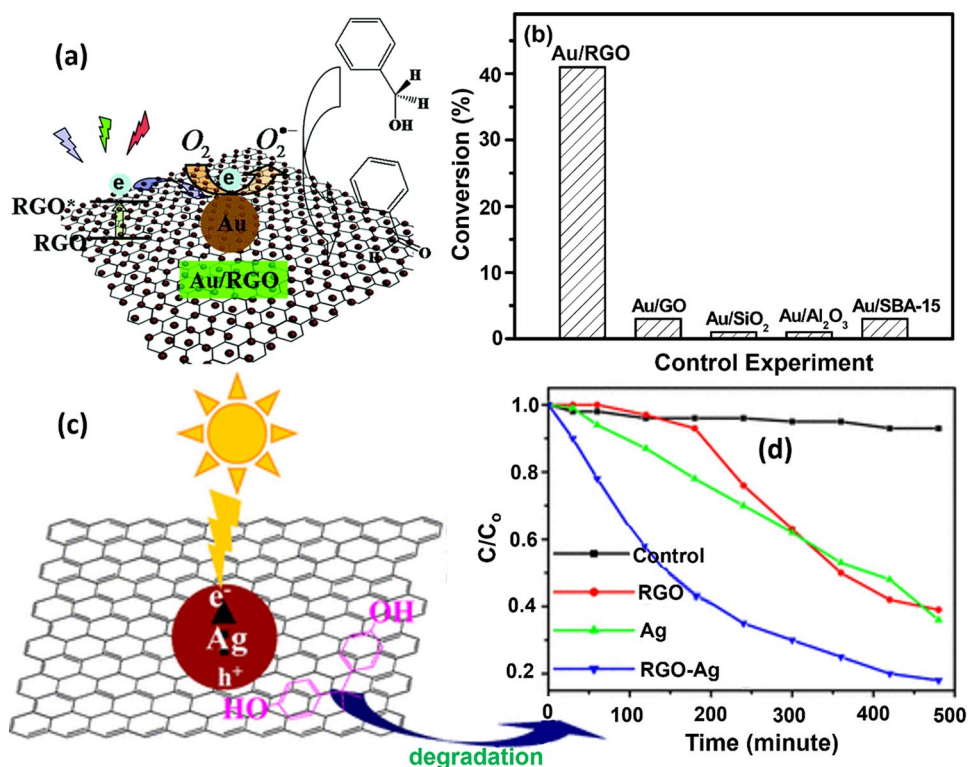
effective surface area of CNTs is lower than that of graphene, reducing their ability to interact with active sites and hindering their photocatalytic performance. Additionally, CNTs suffer from aggregation issues, which further diminish their photocatalytic efficiency.

From the above discussion, it can be concluded that, in comparison to TMDCs, graphene has an edge in terms of high stability and electron mobility, which makes it highly suitable for use in hybrid photocatalysts. Compared to CNTs, although graphene lacks directional charge transport, its 2D platform offers a higher number of interaction sites, thereby enhancing its photocatalytic performance.

Graphene-Based Photocatalyst Systems

As briefly mentioned in the previous section, Graphene essentially acquires the role of a co-catalyst. At the same time, the photoactive element in the nanocomposite system plays the role of light harvester in catalyzing the required reaction. These light-harvesting units of the composite system broadly fall under three headings: Plasmonic metals, Semiconductors (metal oxides and transition metal dichalcogenides), and Organics. Specific articles (Adán-Más and Wei 2013; Prakash 2022; Min et al. 2022) also report the light-sensitive role of graphene where photoexcitation under light irradiation or substrate-mediated electron exchanges such as through dye molecules have also been proved, but such reportage is limited. Graphene-based photocatalyst systems thus can be distinctly categorized as noble metal functionalized graphene, graphene-semiconductors nanocomposites,

Fig. 7 **a** Schematic representation of charge carrier transfer mechanism in noble metal-reduced graphene oxide sheets-based photocatalyst. **b** Bar graph showing the photodecomposition efficiency of Au-RGO for the oxidation of benzyl alcohol. Reproduced with permission (Zhang et al. 2017) copyright 2017 The Royal Society of Chemistry. **c** Schematic represent the efficient charge transfer mechanism between Ag and RGO. **d** Time vs. C/C_0 curves depicting phenol degradation by RGO/Ag/RGO-Ag photocatalyst dissociation. Reproduced with permission (Bhunia and Jana 2014) copyright 2014 American Chemical Society



graphene-sulfide nanocomposites, and graphene-organics photocatalysts (Fig. 6). All these systems have been discussed elaborately in the following section of the review.

Noble Metal-Graphene-Based Nanocomposites

In this section, the integration of plasmonic metal nanoparticles such as Ag and Au with graphene or derivatives of graphene and their employment for efficient water detoxification under light illumination has been discussed in detail. The plasmonic-graphene nanohybrids offer enhanced optical properties due to the surface plasmon resonance effect and provide enhanced effective surface area, improved mobility, and efficient charge separation. The plasmonic-graphene nanohybrids' unique optical absorption, electronic properties, and efficient charge transfer profiles are discussed in detail. Apart from this, several parameters such as photocatalyst amount, scavenger studies, illumination light power, and type of organic pollutant are discussed below.

Conventionally, noble metals nanoparticles in a photocatalysis reaction play dual roles: a) they act as electron sinks that take up photo-charge carriers, extending charge separation efficiency and lifetime, and b) they provide an active reaction surface for chemical reactions to take place by providing lower activation barriers, thereby increasing photocatalyst performance. Additionally, at the nanometer scale, plasmonically active metal particles such as Ag, Au, and Cu have also been utilized as light harvesters owing to

their Surface Plasmon Resonance (SPR) activity upon light irradiation, which is absent in the bulk state (Xie et al. 2020; Shown et al. 2024; Manchala et al. 2019).

When the light frequency with which the metal is illuminated matches the frequency of its surface electrons, an oscillatory restoring force is established, leading to an oscillatory charge density in phase with the falling light. The surface electrons can transfer the gained energy in the form of energetic electrons to the adjoining component, which could be a metal oxide or graphene, GO, or RGO, to drive the photocatalytic reactions. In graphene-metal photocatalysts, metal nanoparticles act as the light-harvesting unit that generates charge carriers in hot electrons due to SPR. In contrast, graphene acts as the co-catalyst, which takes up the electrons and facilitates charge transfer. It has been reported that the different parameters, such as the wavelength of the exposed light and the size of the plasmonic nanoparticles, are strong parameters that influence the charge transfer mechanism of graphene-plasmonic-based nanohybrids (Zhang et al. 2017; Bhunia and Jana 2014). Zhang et al. (2017) used the wet chemical method to synthesize Au-functionalized RGO nanohybrids. The as-prepared Au-RGO sample was employed to decompose benzylic alcohol under Xe lamp exposure. In their study, they have pointed out the contribution of the SPR, thermal, and Au nanoparticle size effects in the photocatalytic efficiency of Au-RGO nanohybrids. Moreover, they have mentioned that the RGO acts as a photosensitizer and creates electrons,

which are further transferred to the Au nanoparticles and decompose the toxic benzyl in alcohol solutions (Fig. 7a). In their photodecomposition studies, 8 mg of photocatalyst samples were employed to convert benzylic alcohols under an Xe lamp source (300 W) (Fig. 7b). Recently, Bhunia et al. (2014) adopted a two-step wet chemical method to synthesize an Ag-RGO nanocomposite system for photo-catalytically degrading phenol-based compounds. An amount of 22 mg of their nanocomposite photocatalyst powder could decompose 100 mg/L solutions of phenol, bisphenol, and atrazine separately. This nanocomposite system could be efficient under UV and visible light sources, possibly due to graphene's UV sensitivity and silver's LSPR activity. The photocatalytic efficiency of their developed Ag-RGO system, when compared with that of bare Ag nanoparticles and RGO, was found to be superior, as observed graphically in Fig. 7c and d. Higher activity of Ag-RGO was ascribed to improved light sensitivity, greater analyte absorption on 2D RGO support, LSPR of Ag nanoparticles, and efficient charge carrier tapping by RGO due to their π conjugate structures, which provide effective charge separation, subsequently improving photocatalytic efficiency. They concluded the role of hydroxyl radicals as the main source for mediating the catalytic dissociation. Although noble metal nanoparticles with single phases bear outstanding SPR properties, these properties can be further improved by creating bi-metallic noble nanoparticles (Darabdhara et al. 2016). In a study conducted by Darabdhara et al. (2016), they synthesized Au-Pd bi-metallic functionalized RGO nanosheets for dissociating aqueous solutions of phenol and its derivatives 2-chlorophenol (2-CP) and 2-nitrophenol (2-NP). They could dissociate a 30 ml solution of phenols within 180–300 min. Their illumination source was natural sunlight. Their study also inferred the role of pH on the dissociation efficiency of the Au-Pd/RGO nanocomposite system. pH of the catalytic reaction was found to affect its efficiency significantly. For phenol and 2-CP, pH 7 was optimal, while for 2-NP, it was 9. Active radicals involved in photocatalytic reactions were majorly identified as hydroxyl radicals. Reduced recombination rates due to metal nanoparticles on the RGO sheets were principally involved in improving their photocatalytic activity. For further development in the field of photocatalysts, nanostructured advanced coating-based photocatalyst is expected to be beneficial for industrial and practical applications. Few research groups reported the plasmonic-graphene-based nanohybrid coating using physical methods and used them for water remediation applications (Birjou et al. 2016; Ren et al. 2015). Recently, Biroju et al. (2016) reported the fabrication and preparation of defective graphene (1 cm $\times$ 1 cm) and graphene oxide (1 cm $\times$ 1 cm)-based substrates using the CVD method combined with the rapid thermal annealing. In the next step, Au nanoparticles were deposited over defective graphene

and graphene oxide-based substrate using the RF-sputtering method. The Au nanoparticles functionalized graphene and graphene oxide-based substrates were employed to decompose MB pollutant molecules solution under visible light illumination. They demonstrated the photodecomposition behavior of Au-graphene and Au-graphene oxide substrates toward the 10 mg/L solution of MB. Their study found that Au nanoparticles, preferably attached to the defect states and oxygenated groups, exist over the surface of graphene. Their study indicated that the efficient charge separation between the Au and defect-rich graphene-based composite majorly contributed to their enhanced photocatalytic activity. In contrast, they mentioned the bi-functionality of Au nanoparticles, which could receive and provide the electrons for RGO and defective graphene, respectively, during the photocatalysis process. In the photocatalytic test, they found 85% efficiency in 120 min of light illumination for the Au-graphene oxide substrates, which is superior to the pure graphene, pure graphene oxide, and their respective plasmonic nanohybrids with Au nanoparticles. Ren et al. (2015) employed a wet chemical method to produce Au-functionalized graphene nanosheets in another study. They used thiophenol to bind Au nanoparticles over the surface of graphene. The fabricated Au-graphene nanohybrids embraced for the decomposition of RhB pollutant molecules solution under visible light (halogen lamp) exposure. In the photocatalytic study, 0.009 mM RhB solution was decomposed using 5 mg Au-graphene nanohybrids in 200 min of halogen lamp exposure. Using the laser-assisted photolysis method, Chen et al. (2020b) fabricated the highly stabilized Au-graphene and Au-graphene oxide nanohybrids. They have employed the prepared nanohybrids (5 mg) for the photodecomposition of MB molecules solution (10 mg/L) under visible, UV, and solar light. They have concluded that the maximum photocatalytic activity was achieved under solar light for Au-graphene (94.5%) and Au-graphene oxide (92.1%)-based substrates for the decomposition of MB solution.

From the above studies, it can be concluded that plasmonic nanoparticles functionalized graphene and its derivatives can efficiently decompose various organic pollutants due to enhanced optical absorption, effective charge separation, high numbers of adsorption sites, and improved stability. Thus, plasmonic-graphene composite has been proven as a promising candidate for water detoxification and several photocatalysis processes. On the other hand, several challenges hinder the development of plasmonic-graphene nanohybrids for practical applications. Since noble metal nanoparticles (Ag, Au) are not cost-effective, they restrict the bulk usage of plasmonic-graphene composite in different industrial and practical applications. Apart from this, other challenges, such as environmental risk, UV light activation, and reusability, have to be resolved for further progress in the field of plasmonic-graphene-based photocatalysis.

Semiconductor Metal Oxide-Graphene-Based Nanocomposites

For the sake of cost-effectiveness in the graphene-based photocatalyst, the attachment of different semiconductors such as TiO_2 , ZnO , and CuO with graphene and their derivatives has been proposed. This section presents an overview and detailed discussion for metal oxide-graphene based. In addition, their employment in water treatment using advanced oxidation processes has been highlighted. These metal oxide-graphene nanocomposites provided improved optical properties and efficient charge separation. According to the optical absorption requirements, different semiconductors (for UV light TiO_2 and visible light CuO) can be chosen to enhance the optical properties and create a synergistic effect among graphene and metal oxide. The unique optical absorption, electronic characteristics, and synergistic effect of metal oxide-graphene nanocomposite are elucidated. Apart from this, this section also highlights several advantages and challenges of the different metal oxide-graphene nanocomposites-based systems in detail.

Semiconductor metal oxides with requisite bandgaps are well-known photocatalysts often complexed with graphene or its derivatives to form photocatalysis-active graphene-metal oxide composites. Unlike metal-graphene systems, the photocatalytic activity in metal oxide-graphene composites relies on the electronic bandgap in the photocatalyst (Hong et al. 2020; Sharma et al. 2022; Pervez et al. 2021; Zhu et al. 2020; Song et al. 2019a, b). Upon light irradiation on a metal oxide, it absorbs energy (photons) matching or exceeding the difference between its valence and conduction band, also known as the bandgap. The gained excess energy pushes the charge carriers, i.e., electrons, from the valence to the conduction band, leaving a vacancy or a hole in the valence band. These charge carriers, on separation, migrate to the metal oxide surfaces, which are enriched with catalytically active reaction sites. They participate in oxidation–reduction reactions at the surface to generate reactive radical species that attack the toxin molecules, leading to their disintegration (Singh et al. 2020f, g). However, the efficiency of a semiconductor metal oxide photocatalyst is generally subdued because the rate of recombination of the generated hole and electron is much higher than the rate at which they can catalyze surface reactions (Goktas et al. 2022b; Aslan et al. 2024). Improving charge separation can thus fundamentally help us enhance the photolytic efficiency of a system. Graphene, as adequate conductive support with low Fermi energy, can modulate the photocatalytic efficiency in the composite by acting as an electron sink (Merino-Díez et al. 2017). Graphene acts as a shuttler co-catalyst which accepts electrons from the metal oxide bandgap retarding their union with a hole, subsequently resulting in improved photocatalytic activity by allowing increased participation

in photo-redox reactions (Burkholder et al. 2022; Pastrana-Martinez et al. 2014). Consider the following examples from the literature for a detailed understanding.

As we have discussed above, to create the UV light-sensitive photocatalyst, graphene or its derivatives are integrated with wide bandgap semiconductors such as TiO_2 , ZnO , and NiO . The photocatalytic for several types of organic pollutants and the effect of different light exposures on this system have been explored briefly using the studies mentioned below. Pastrana-Martinez et al. (2014) embraced the joint effort of the liquid-phased deposition method with the thermal reduction method to fabricate GO/TiO_2 nanohybrids. They have varied GO content with a fixed amount of TiO_2 nanostructures and optimized the GO content in GO/TiO_2 nanohybrids for enhanced photodecomposition activity. In their photocatalytic experiment, textile waste molecules (methyl orange) and a pharmaceutical waste molecule [diphenhydramine (DP)] were decomposed under exposure to UV light as well as visible light. They have discussed that under visible light exposure, the graphene gets activated and injects the photoexcited electrons in the CB of the TiO_2 nanostructures; consequently, the concentration of the electrons was increased in the CB of TiO_2 . Conversely, TiO_2 nanostructures sequentially generate electrons and holes in their CB and VB under UV light exposure. In such cases, graphene acts as an electron scavenger and reduces the recombination rate in TiO_2 . In both cases, the photocatalytic activity of GO/TiO_2 is found to be enhanced. Cho et al. (2015) used hydrothermal and Hummers' methods to successfully synthesize a $\text{TiO}_2/\text{Graphene}$ nanocomposite system. Figure 8a, b represents their TEM and HRTEM characterization, which gives a clear picture of the nanocomposite composition. As observed, mesoporous TiO_2 with near spherical morphology was found interspersed on the 2D-graphene support. The composite formation was confirmed by estimating the spacing of their HRTEM. The photocatalytic activity of their synthesized nanocomposite was compared to the commercially available P25, RGO-P25, and 2D bare TiO_2 . Expectantly synthesized TiO_2 -RGO demonstrated far higher photocatalytic activity than the rest Fig. 8c and d. A 30 μM methylene blue dye solution was degraded within 80 min when exposed to an Xe lamp. The photocatalyst concentration for the study was 10 mg/30 ml. Co-catalytic activity of reduced graphene was ascribed as a probable reason for its improved activity.

In another study, Sawant et al. (2015) complexed wide bandgap semiconductor ZnO with graphene to form a composite. Since ZnO is known to be an excellent UV absorber, the decomposition study was conducted in the presence of UV light, where Rhodamine B and Methyl Orange dyes were used as model toxin analytes. These electrolytically exfoliated ZnO nanosheets composite with graphene could effectively decompose 96% and 89% of 10 ppm RhB and MO

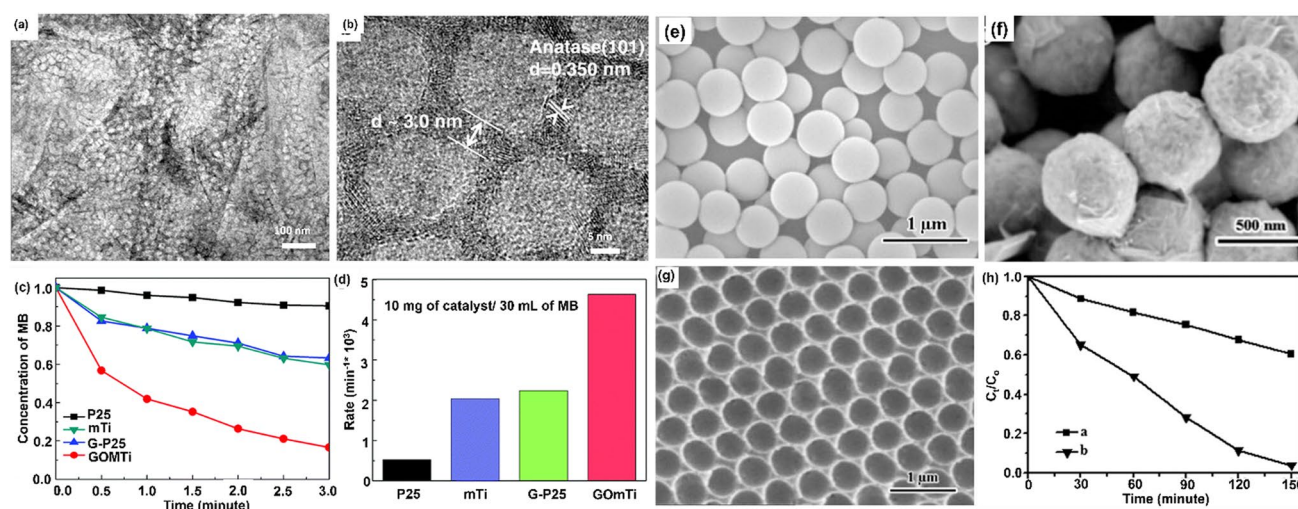


Fig. 8 **a** and **b** TEM and HRTEM image of 2D TiO₂ mesoporous/RGO nanocomposites, respectively, **c** photocatalytic decomposition of MB dye solution using p25, RGO-P25, 2DTiO₂, and 2DTiO₂/RGO, respectively, **d** bar graph reveals the rate kinetics of p25, RGO-P25, 2DTiO₂, and 2DTiO₂/RGO. Reproduced with permission (Cho et al. 2015) copyright Royal Society of Chemistry 2015,

e FESEM image of cationic monodisperse sphere, **f** FESEM image of graphene—cationic monodispersed spheres, **g** FESEM image of GO/TiO₂ mesoporous nanocomposite, **h** photocatalytic degradation of MB dye solution using p25 and GO/TiO₂ mesoporous nanocomposite. It is reproduced with permission (Yang et al. 2016) copyright 2016 Elsevier

dye solutions within 420 and 240 min, respectively. Their study concluded the role of optimum graphene loading in tuning the lifetime of photogenerated charge carriers, which plays a significant role in retarding recombination. Recently, Yang and his co-workers (Yang et al. 2016) reported the template-mediated fabrication of TiO₂-graphene nanocomposites capable of photo-catalyzing methylene blue dye under a Xenon lamp. Their study showed that with 1 mg of the photocatalyst material, 10 mg/L dye solution could be completely dissociated within 150 min. They further performed several studies with scavengers to understand the role of active radicals in improved photocatalysis. Scavenger studies concluded that superoxide ($\cdot\text{O}_2$) is the leading free radical responsible for enhancing photolytic efficiency. As understood in the introduction section, they also proposed the co-catalyst role of graphene as a charge separator to be the leading cause for improving photocatalytic activity. Their synthesized TiO₂/mesoporous graphene exhibited 6.5 times better activity when compared to commercially available p25. Figure 8e–g represents the microscopic characterization of the synthesized TiO₂/graphene complex, while Fig. 8h shows the comparative photocatalytic efficiency data between the complex and p25. The primary idea behind sharing these examples is to make the readers understand the diversity of materials and methods used to synthesize these photocatalysts. The choice of photocatalyst material may vary depending on your application needs and toxin type. So far, more than 100 such types of graphene-semiconductors photocatalysts have been produced and studied for targeting a variety of toxins, including dyes, organic compounds, and

even biological contaminants such as bacteria. A few such examples are discussed below, while a more cumulative article list is summarized in Table 1.

Arshad et al. (2017) used a less common yet highly effective semiconductor NiO with graphene to decompose the anionic methyl orange dye using sunlight as the renewable light source. With 0.2 g/L photocatalyst concentration, 20 mg/L dye solution could be dissociated efficiently

Upon experimental exploration of the photocatalysis mechanism, they proposed improved conductivity in graphene as a fundamental contributor to enhanced photocatalysis. The photogenerated electrons could be transferred to graphene nanostructures, leaving unpaired holes behind. These could then effectively increase toxin disintegration by catalyzing photo-redox reactions. Figure 9a and b presents the TEM image of NiO nanostructures and Graphene/NiO nanostructures. In contrast, Fig. 9c and d depicts their photocatalytic activity and photodegrading rate kinetics curves, indicating improved photocatalytic activity of Graphene/NiO nanostructures compared to pure NiO nanostructures.

Integrating visible light active metal oxide like CuO, Ag₃PO₄, and Bi₂WO₆ with graphene or its derivatives can provide unique optical properties and enhanced energy harvesting capability. In another study, RGO-CuO was fabricated by Aroob and his co-workers (Aroob et al. 2021) by combining two different methods: Hummer's and biosynthesis wet chemical method. When exposed to solar light, their synthesized RGO-CuO nanocomposite could decompose five classes of organic dyes: Methylene Blue, Methylene violet, Methyl Green, Rhodamine B, and Methyl Orange. In

Table 1 Summary of some recent publications on bi/multilayer graphene-based photocatalyst systems

No	Photo catalyst and type	Synthesis methodology	Morphology	Dye/organic component degraded	Degradation (%)	Amount of catalyst	Degradation time (min)	Light source	References
1	Au-GO/BiVO ₄ -ZnO	Wet chemical method with sonication process	BiVO ₄ -ZnO particles RGO sheets Au Np	MB 10 µM	98	10 mg/mL	60	Sunlight	(Kumar et al. 2023)
2	ZnO-RGO (Binary)	Chemical precipitation method	ZnO nanoparticles/graphene sheets	MB 10 µM	100	50mg	100	Xenon lamp	Gayathri et al. (2014)
3	ZnO-rGO-Au (Ternary)	Combination of thermal treatment photoreduction	ZnO nanorods covered with GO and spherical AuNps	R6G 10 ⁻⁵ M	Conc.-time dependence	1000 µg/mL	32	UV lamp	Wen et al. (2013)
4	Ag/AgBr/Graphene Oxide Nanocomposite	Surfactant assisted chemical method and micro-emulsion method	Ag/AgBr nanoparticles on GO sheets	MO 4 ml solution of 20mg/L	Conc.-time dependence	7 mg	20	Sun Light	Zhu et al. (2012a)
5	Nitrogen-doped graphene/CuO nanocomposites	Chemical etching with hydrothermal method	CuO nanowires on graphene sheets	MO 10mg/ml	100	10 mm × 10 mm × 1 mm	35	Xenon lamp	Zhang et al. (2014)
6	Au/Graphene/C ₃ N ₄ nanocomposites	Chemical method + annealing (550°C for 3 h)	Au nanoparticles/Graphene sheets/C ₃ N ₄ sheet	MB/CIP 10mg/L	100	100 mg	240	Xenon lamp	Xue et al. (2015)
7	Cu-RGO Nanocomposites	Chemically exfoliated + chemical method	Cu modified on RGO sheets	RhB 5.3 × 10 ⁻³ mM	100	6.3mg	172	Xenon lamp	Xiong et al. (2011)
8	Ag/TiO ₂ /graphene nanocomposite	Hydrothermal Method	Ag nanoparticles/TiO ₂ nanoparticles/graphene sheets	MB 10mg/L	100	100 mg	60	Halogen lamp	Wen et al. (2011)
9	Cu ₂ O/TiO ₂ /Graphene nanocomposites	Chemical reduction method	Cu ₂ O nanowire/TiO ₂ /nanowire/graphene sheets	MO, Benzoic Acid, Aniline (10ppm)	98	rGO/Cu ₂ O/TiO ₂ membrane	120	Xenon lamp	Wang et al. (2018a, b)
10	Graphene-V ₂ O ₅	Hummer's method + hydrothermal method	V ₂ O ₅ nanorod/graphene sheets	MB 10mg/L	100	10 mg	150	UV light Visible light Sunlight	Shannugam et al. (2015)
11	TiO ₂ QDs/Graphene	Wet chemical method	TiO ₂ QDs/graphene sheets	PFOA	100	4mg/200mL	600	Hg lamp	Pervez et al. (2021)
12	MnO ₂ /GR nanocomposites	Hummer's method + hydrothermal method	MnO ₂ NPs/Graphene	Tetracycline	99.4	-	-	-	Song et al. (2019a, b)

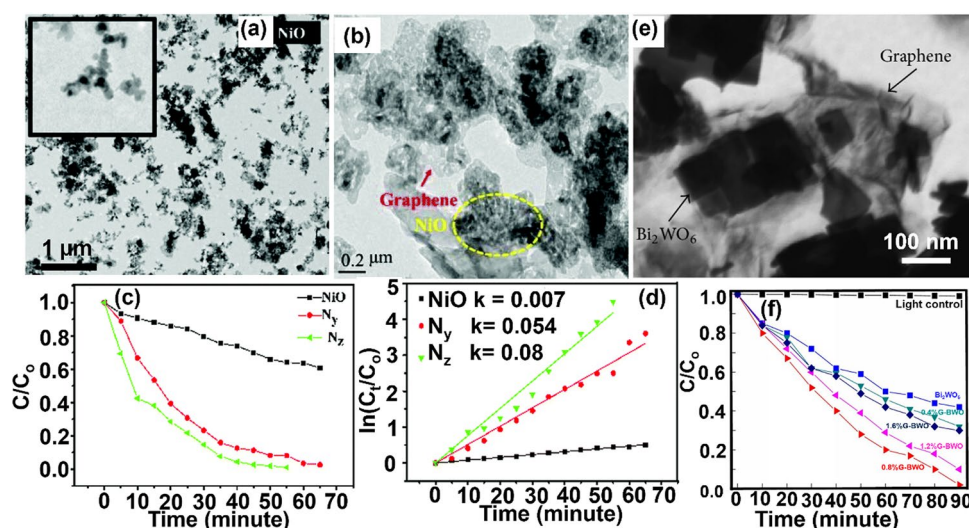


Fig. 9 **a** TEM image of NiO nanostructures, **b** TEM image of Graphene/NiO nanostructures, **c** photocatalytic degradation of MB dye using as-prepared photocatalyst, **d** linear fitted $\ln(C/C_0)$ vs exposure time graph for the estimation of rate constant. Reproduced with permission (Arshad et al. 2017) copyright 2017 Royal Society

of Chemistry, **e** TEM image of water-soluble graphene nanosheets, **f** Photocatalytic degradation of RhB dye solution using Bi_2WO_6 attached graphene-based photocatalyst. Reproduced with permission (Hu et al. 2016) copyright 2016 Wiley

their photocatalytic experiment, 5 mg photocatalyst material decomposed 10 ppm solution of MB, MV, MG, MO, and RhB dye solution separately in 60 min. In their photocatalytic mechanism, they highlight that under sunlight exposure, the electron-hole pair was created in the CB and VB of CuO nanostructures, which were further captured by the graphene oxide, which gave rise to the reduction in the recombination rate in CuO nanostructures.

Hu et al. (2016) used the hydrothermal method to fabricate Bi_2WO_6 attached graphene nanohybrids (Fig. 9e). These Bi_2WO_6 functionalized graphene sheets have been applied to degrade Rhodamine B dye solution in water using visible light. The Bi_2WO_6 attached graphene nanosheets exhibit boosted photocatalytic degradation rates compared to pristine Bi_2WO_6 nanostructures (Fig. 9e). The higher photocatalytic activity of Bi_2WO_6 /graphene was ascribed to the high adsorption capability of graphene sheets and efficient charge transfer among the Bi_2WO_6 and graphene. They also demonstrated the effect of the pH values and amount of photocatalyst on the efficiency of Bi_2WO_6 /graphene. Figure 9f presents its characterization and reaction kinetics plots.

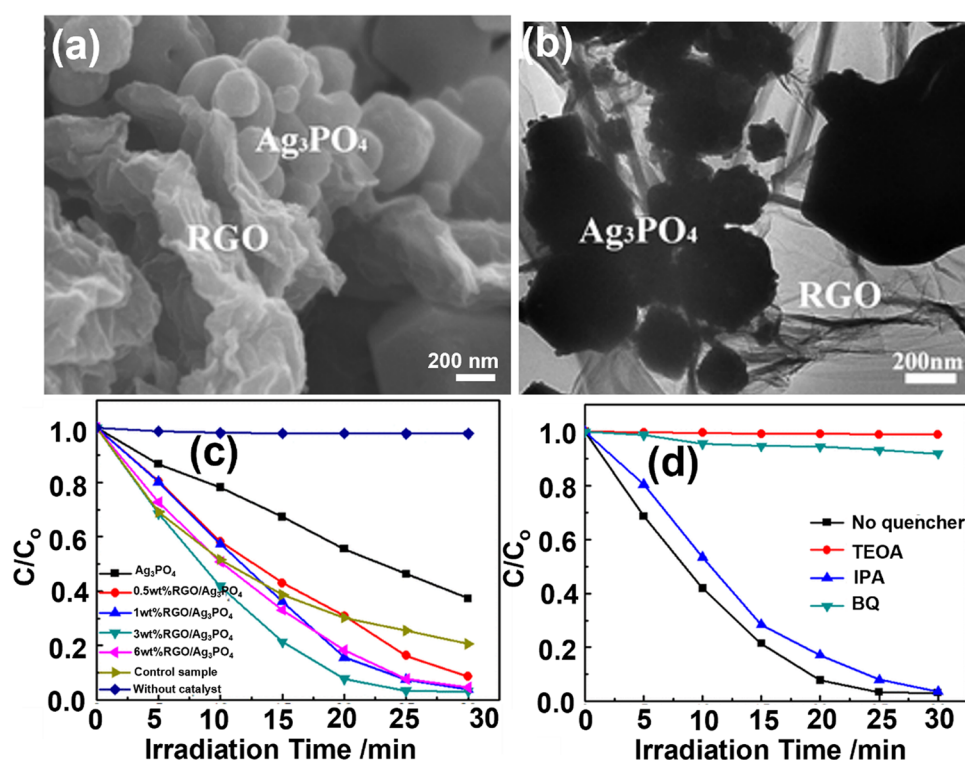
Chai et al. (2014) fabricated a novel Ag_3PO_4 -Graphene nanocomposite using a two-step step-wet chemical method and studied their photodegradation behavior under visible light illumination. The TEM images of the Ag_3PO_4 -Graphene nanocomposites are depicted in Fig. 10a and b. Using 50 mg Ag_3PO_4 -Graphene nanocomposite, 10 μM RhB dye solution was degraded. In their study, they also varied the loading of Ag_3PO_4 on graphene sheets to optimize the effective Ag_3PO_4 loading and maximize

photodegradation (Fig. 10c). Additionally, their scavenger tests concluded the superior role of holes and superoxide radicals in affecting their photodecomposition efficiency (Fig. 10d).

In addition to single semiconductor-graphene composites, a combination of semiconductor-graphene with an additional functional component such as metal nanoparticles, another second semiconductor or organics have also been synthesized. Such multinary composites often act as light harvesters while graphene channelizes electron flow. The following section illustrates some examples of military photocatalysts.

Ghasemi and his co-workers followed a three-step wet chemical route to synthesize an Au-graphene double-functionalized TiO_2 nanocomposite system and used it to decompose Acid blue 92 (Ghasemi et al. 2017). 80 ppm of the photocatalyst material could completely dissociate 20 ppm of the dye solution. The excitation source was a mercury lamp, while the stipulated time for the study was 120 min. Compared to their control samples, the catalytic efficiency of their bi-functionalized Au-graphene- TiO_2 nanocomposite was higher. The reasons identified for the same were the SPR effect, bandgap narrowing, enhanced surface area, and reduction in the recombination rates. In another study, Roy et al. synthesized a bi-functional Au/ZnO/Graphene nanocomposite for degrading nitrobenzene (Roy et al. 2013). This composite could dissociate 50 ml, 5 mM nitrobenzene solution within 140 min under UV light illumination. The catalyst load was in mg/ml. TEM images of the synthesized nanocomposite are represented by Fig. 11a, while Fig. 11b represents its degradation kinetics as a function of time.

Fig. 10 **a** and **b** TEM image of Ag_3PO_4 -Graphene nanocomposite (3% wt), **c** photodegradation behavior of RhB dye solution by using pure Ag_3PO_4 and different loading of Ag_3PO_4 on graphene nanosheets. **d** A scavenger experiment revealing the responsible radicals for photocatalytic behavior under visible light exposure. Reproduced with permission (Chai et al. 2014) copyright 2014 American Chemical Society



Both Au and Graphene act as an electron sink, scavenging electrons upon excitation, leading to retarded recombination and, thus, better catalytic activity.

Besides Au, silver nanostructures have also been used to functionalize graphene and its nanocomposites, further improving its photocatalytic efficiency. Silver nanoparticles have visible region LSPR, show excellent catalytic properties, and are cost-effective. Liu et al. (2018) synthesized a set of $\text{Ag}/\text{TiO}_2/\text{Graphene}$ nanocomposite membranes with different Ag loadings to optimize the Ag content for maximal activity. Ag content on graphene sheets varied between 1 and 7 wt%. Compared to the control, the photocatalytic activity upon Ag functionalization showed drastic improvement in MB dissociation with increasing Ag content. 10 mg/L MB dye solution was degraded in 170 min under xenon light irradiation. They have also concluded that the high adsorption capability of graphene, SPR properties, and the synergetic effect of Ag nanoparticles are responsible for the enhanced photocatalytic activity of Ag doping on the Graphene/ TiO_2 nanocomposites. Figure 11c and d reveals the existence of TiO_2 nanorods and Ag nanoparticles on the graphene sheet. At the same time, Fig. 11e–g depicts the photocatalytic degradation of MB dye with the variation in TiO_2 loading, Ag loading, and different as-prepared photocatalyst, respectively.

The studies mentioned above and their explanation highlight the attractive strategies to design the metal oxide/graphene-based nanohybrids and elaborate their efficient charge transfer mechanism for boosting the photodecomposition

ability for different pollutants. Apart from this, several parameters like reusability, pH variations, and photocatalyst dosage are discussed well for different designs of metal oxide/graphene-based. Thus, metal oxide-graphene nanohybrids have been proven to be promising alternative cost-effective photocatalyst materials. It offers enhanced photodecomposition performance with various pollutant molecules owing to their efficient charge transfer mechanism, enhanced reactive sites, and improved light-harvesting capabilities. However, if we talk about the dark side, the metal oxide-graphene nanohybrid photocatalyst suffers from stability and scalability for commercial usage and harmful environmental effects. Apart from this, there are some challenges to developing metal oxide-graphene nanohybrids, such as complex synthesis methods, which give rise to variations in their performance.

2D-Sulfides-Based Graphene Nanocomposites

Although plasmonic-graphene nanohybrids and semiconductor-graphene nanocomposites exhibit remarkable photodecomposition ability, their activity encountered different limitations. For example, plasmonic-graphene nanohybrids-based photocatalysts bear enhanced activity due to the surface plasmon resonance effect. However, the limited stability and dispersion of plasmonic nanoparticles are the main hurdles to further developing plasmonic-graphene-based photocatalysts. On the other hand, the photocatalytic efficiency of semiconductor-graphene nanocomposite is hindered due to

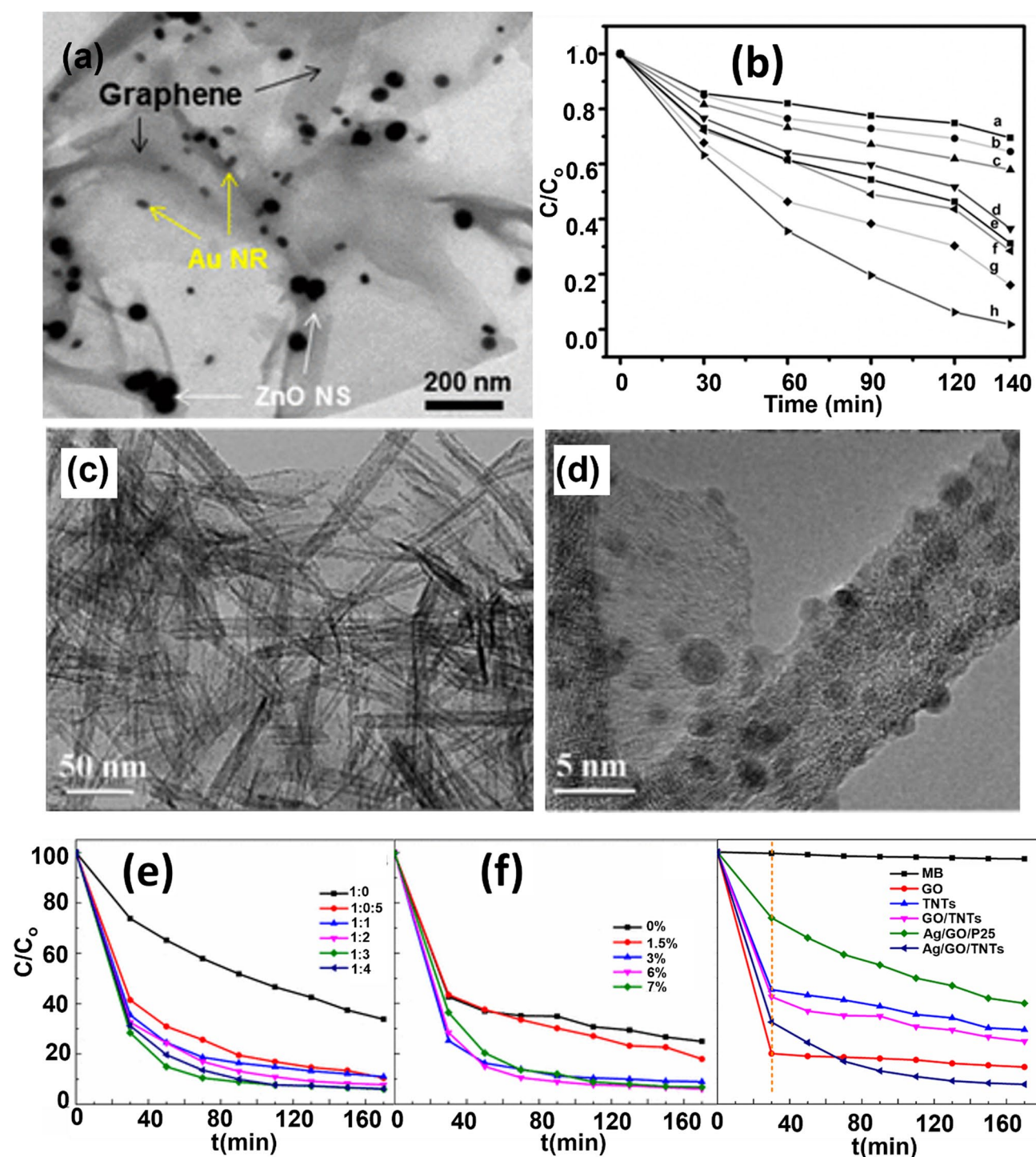


Fig. 11 **a** TEM image of graphene-ZnO-Au nanocomposite, **b** time kinetics of nitrobenzene degradation for **a** control, **b** graphene (0.5 mg/ml), **c** Au nanorods (0.5 mM), **d** ZnO nanosheets (18 mM), **e** commercial photocatalyst P25 (18 mM), **f** graphene-Au nanorod (4 mg/mL), **g** graphene-ZnO nanosheet (4 mg mL⁻¹), and **h** graphene-ZnO-Au nanocomposites (4 mg/mL). Reproduced with permission (Roy et al. 2013) copyright 2013 American Chemical Society, **c** TEM image revealing Ag-doped Graphene/TiO₂ nanocomposites, **d** higher

magnification view of Ag-doped Graphene/TiO₂ nanocomposites, **e** variation in the photocatalytic degradation of MB dye by varied the TiO₂ nanorod loading, **f** changes in the photocatalytic decomposition of MB dye by varied the Ag loading, **g** photocatalytic degradation of MB dye using as-synthesized photocatalyst (GO, TNT, GO/TNT, Ag/GO/P25 and Ag/GO/TNT). Reproduced with permission (Liu et al. 2018) copyright 2018 American Chemical Society

its recombination rate and mismatch in band alignment. To overcome this limitation, nanocomposites of graphene with 2D-layered sulfides are one of the advanced approaches. Recently, sulfide-based, 2D-layered nanostructures aroused great attention in photocatalysis due to their outstanding optical and effective surface area properties (Tumbul et al. 2024; Aslan et al. 2023; Goktas et al. 2022a; Aba et al. 2024). The attachment of 2D-layered sulfides offers synergistic interaction and enhanced electron coupling with an adequate surface area, which can significantly provide new insight for developing graphene-based photocatalysts for practical applications.

Recently, the integration of graphene with two-dimensional sulfides such as MoS_2 , WS_2 , In_2S_3 , and SnS_2 , etc., has come into the limelight which was found effectively beneficial for the use in various environmental and energy generation applications (Shah et al. 2021; Yang and Xu 2013b; Chauhan et al. 2016). As mentioned above, graphene with honeycomb structures exhibits outstanding physical properties. On the other hand, due to the zero bandgap, graphene suffers from lower catalytic activities. In addition, due to low surface area and zero bandgap, the applicability of graphene is reduced for various optoelectronic applications (Li et al. 2016). Moreover, graphene and its derivatives reveal the optical absorption in UV light, which further limits its performance in environmental and optoelectronic applications (Rosman et al. 2018). With the formation of graphene nanocomposites with 2D-layered sulfides, their effective surface area was enhanced significantly. Secondly, 2D-layered sulfides mainly exhibit a narrow bandgap and absorb the visible light in the solar spectrum. Thus, integrating 2D-layered sulfides with graphene enhances the optical region, further improving their performances for several energy generations and environmental and energy storage applications (Wang and Long 2021). With the attachments of the 2D-layered nanostructures (for example, MoS_2) with the graphene, effective charge transfer occurs, remarkably enhancing their capability for different photocatalytic applications. Under light excitation, the electron holes were generated in the CB and VB of the 2D-layered nanostructure. Here, the graphene attracts the electrons from the CB of the 2D-layered nanostructures and reduces the recombination rate in 2D-layered nanostructures; consequently, efficient charge separation occurs (Zhang et al. 2016; Chakrabarty et al. 2018; Ali et al. 2024). Thus, combining 2D-graphene sheets with the TMDCs is expected to provide outstanding photodecomposition behavior due to their excellent adsorption capabilities, efficient charge transfer mechanism, and modified optical profiles.

Zhang et al. (2016) prepared the heterojunction among the RGO and MoS_2 nanostructures using the hydrothermal method and employed it as a photocatalyst to remove the RhB molecule. Their study demonstrated that the RGO

content on MoS_2 nanostructures increased significantly with the increment of the photodecomposition process. Their studies indicate that the MoS_2/RGO nanocomposite exhibited 2.5 times higher photodegradation efficiency than pristine MoS_2 . In their photocatalysis studies, 10 mg of prepared photocatalyst was used to decompose 20 mg/L of RhB molecules solution for 180 min under visible light exposure (1 kW, Xe lamp). It has been shown that heterojunction among graphene and TMDCs can further enhance photocatalytic activity when combined with other semiconductors (Chakrabarty et al. 2018). Chakrabarty et al. (2018) reported the synthesis of $\text{RGO-MoS}_2/\text{NiCo}_2\text{O}_4$ nanocomposites by using two-steps hydrothermal method and applied this nanocomposite for degradation of Rhodamine B dye under visible light irradiation. Figure 12a and b indicates the TEM and HRTEM images of the RGO-

$\text{MoS}_2/\text{NiCo}_2\text{O}_4$ ensure their formation. In their photocatalytic experiment, 95% rhodamine B dye solution was decomposed in 70 min under visible light exposure (Fig. 12c, d). They proposed the presence of RGO photogenerated electrons efficiently maintaining the flow between NiCo_2O_4 and MoS_2 as the driving force behind improved photocatalysis. Apart from this, RGO also helps.

to minimize the recombination rate and enhance the photocatalytic activity. They also concluded that hydroxyl radicals essentially affect the photocatalytic degradation activity of rhodamine b (RhB) dye under visible light illumination. Recently, Nasr et al. (2022) reported the formation of RGO/WS_2 nanocomposites by using the combined effort of the chemical vapor deposition method with the ball milling process. Their study varied the graphene content over WS_2 nanostructures and optimized them for enhanced photocatalytic water-spitting application. In their photocatalytic water splitting experiment, a sample prepared with 1% graphene/ WS_2 reveals the enhanced photocurrent density of $95 \mu\text{A}/\text{cm}^2$, superior to the pristine WS_2 samples. They highlighted that a sample prepared with 1% graphene/ WS_2 was found to have 3.3 times better performance than pristine WS_2 nanoflakes. They have also pointed out that enhanced photocatalytic performance occurs due to the efficient charge transfer among graphene and WS_2 nanostructures.

Besides the TMDCs, graphene and its derivatives showed improved photodecomposition ability with other 2D-layered sulfides such as In_2S_3 and SnS_2 . The combination of 2D-layer materials with graphene not only provided additional optical absorption but also tremendously increased the number of active sites. Moreover, since graphene and 2D-layered sulfides are dangling bonds and layered structures, their attachment is comparatively more straightforward than other graphene-based reported photocatalysts. Yang and Xu (2013b) used the self-assembly method to fabricate the $\text{In}_2\text{S}_3/\text{graphene}$ nanocomposites and employed them to decompose the 4-Nitroaniline solution under visible

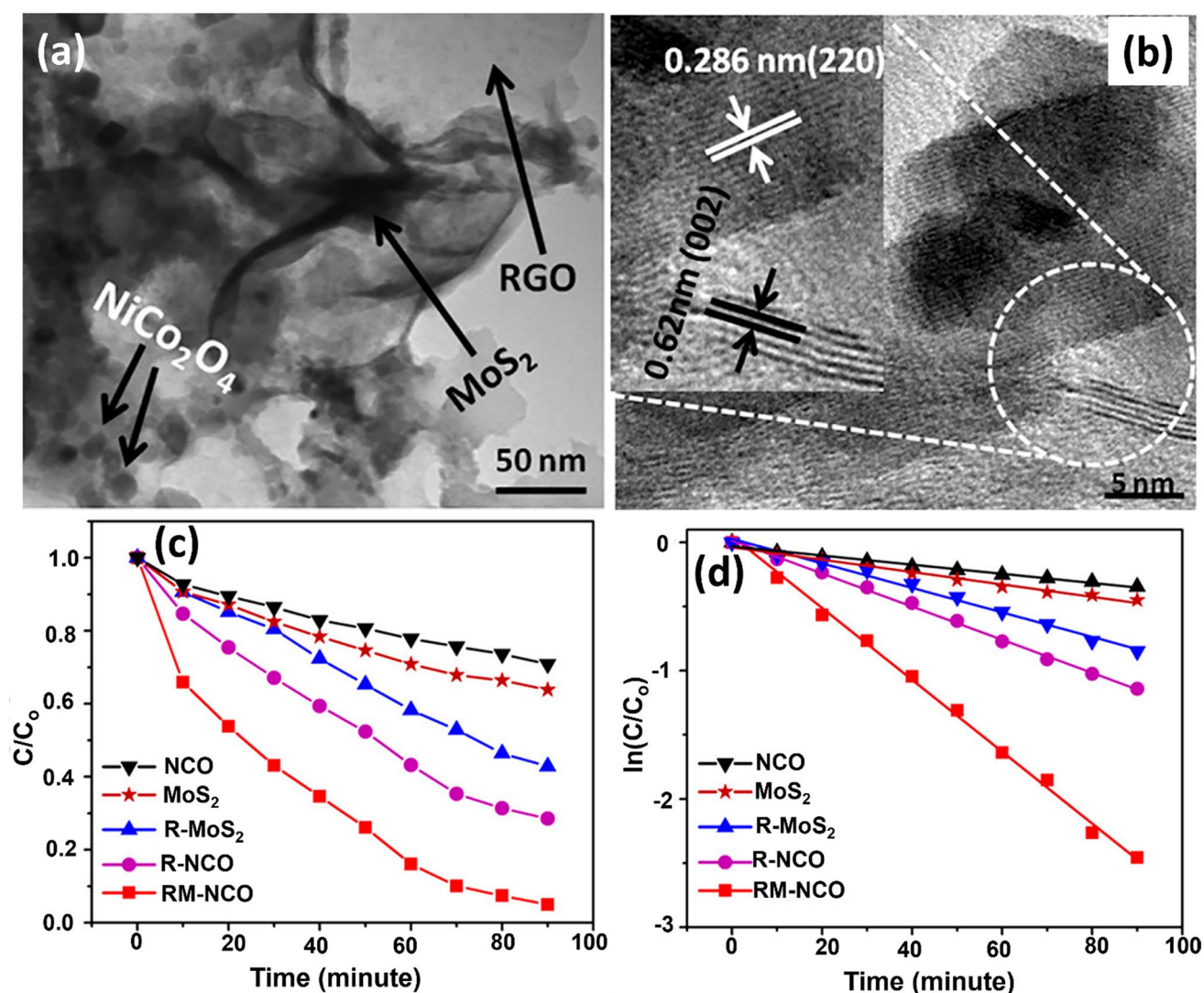


Fig. 12 **a** TEM image of RGO-MoS₂/NiCo₂O₄ nanocomposites, **b** HRTEM image of RGO-MoS₂/NiCo₂O₄ nanocomposites, **c** photocatalytic degradation of RhB dye using pristine sample and RGO-MoS₂/NiCo₂O₄ nanocomposites, **d** rate constant estimation of different

photocatalysts such as NiCo₂O₄, MoS₂, RGO-MoS₂, RGO-NiCo₂O₄ and RGO-MoS₂/NiCo₂O₄. Reproduced with permission (Chakrabarty et al. 2018) copyright 2018 American Chemical Society

light exposure. They have varied the graphene content on the surface of the In₂S₃ nanostructures and obtained the optimum graphene content for superior photodecomposition behavior. In the photocatalytic studies, a 10 ppm solution of 4-Nitroaniline was decomposed using 40 mg of the In₂S₃/graphene nanocomposites for 120 min under visible light illumination (Xe lamp- 300 W). They highlighted the chemical attachment among the graphene and In₂S₃ nanostructures, which is responsible for the efficient charge transfer and enhanced conductivity of graphene and In₂S₃ nanostructures. Due to the improved charge separation, the lifetime of the charge carriers significantly increased; consequently, the photodecomposition activity of graphene/In₂S₃ was enhanced remarkably. Figure 13a shows the efficient

charge transfer process due to the photocatalysis process. At the same time, the decomposition profiles of 4-nitroaniline solutions using different pristine In₂S₃ and graphene/In₂S₃ nanocomposites are presented in Fig. 13b.

An et al. (2013) employed a one-pot hydrothermal method to prepare In₂S₃ sheets attached to graphene nanocomposites. They used a visible-light-induced photocatalytic process to remove organic pollutants and Cr ions from the wastewater. Their study used different sulfide sources (thiourea, thioacetamide, and cysteine) to prepare In₂S₃ nanosheets. They optimized that the sample prepared with cysteine exhibited superior photodecomposition activity under visible light. In photocatalytic studies, they demonstrated that the 20 mg of photocatalyst sample decomposed the 25 ppm MO solution

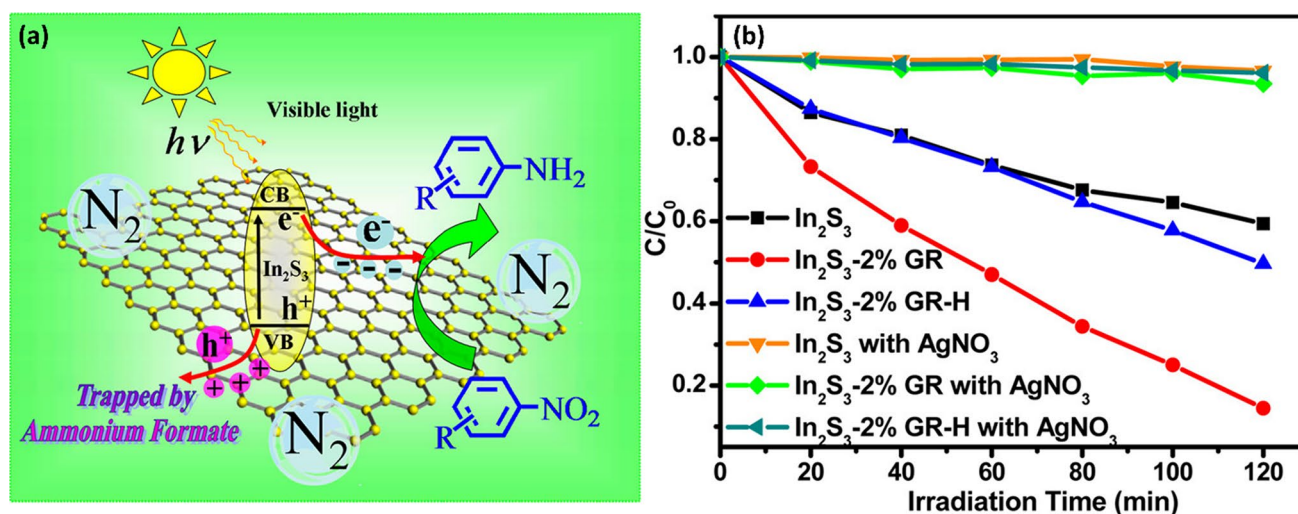


Fig. 13 **a** Pictorial view indicates the efficient charge transfer among the In₂S₃ and graphene during the photocatalytic process, **b** photodecomposition of 4-nitrosamine using the different In₂S₃-graphene-

based nanocomposites. Reproduced with permission (Yang and Xu 2013b) copyright 2013 American Chemical Society

under visible light exposure (Xe, 300 W). A 10 mg photocatalyst sample was employed to dissociate 10 mg/L solution in 120 min of light exposure for the removal studies of Cr ions. They also showed that prepared In₂S₃-graphene nanocomposite exhibited superior photocatalytic activity to pristine In₂S₃ sheets. The improved photodecomposition activity could be assigned to efficient charge transfer among the In₂S₃ sheets and graphene, enhanced optical absorption, and effective surface area due to the 2D-layered structures of In₂S₃ and graphene.

Recently, Chauhan et al. (2016) used a hydrothermal process for the fabrication of SnS₂/graphene nanohybrids and embraced them for the conversion of NB to aniline and photodecomposition of RhB, MB, and removal of Cr ions from the wastewater under visible light illumination. They have attached the SnS₂ nanoparticles over the surface of graphene sheets. Their study mentioned that the SnS₂/graphene sample indicates superior photodecomposition ability compared to pristine SnS₂ and graphene. In their photocatalytic experiment, 12.5 mg of each prepared photocatalyst sample was used to decompose 2 mM of NB, 0.01 mM of MB, 0.01 mM of RhB, and 0.5 mM of Cr solution under visible light exposure. The exposure time for NB and Cr solution conversion is 90 min, while the exposure time for the RhB and MB were 15 and 5 min, respectively. The improved photocatalytic behavior has been ascribed to the enhanced effective surface area, existence of functional groups, and effective charge separation among SnS₂ and graphene. Dashairya et al. (2019) employed hydrothermal methods to prepare SnS₂ QDs functionalized graphene oxide sheets and used them to decompose organic pollutants under visible light illumination. Their study mentioned that

effective bandgap narrowing improves the photocatalytic efficiency of SnS₂-attached graphene oxide sheets. Moreover, the enhanced surface area of the SnS₂/graphene oxide composites also plays a key role in increasing their interaction with the pollutant molecules. In the photocatalytic test, they decomposed the RBB and RBR pollutant molecules using a synthesized photocatalyst sample for 150 min under visible light illumination. They highlighted that SnS₂/graphene oxide composites exhibited superior photodegradation performance compared to pristine SnS₂.

The current summarized sulfides-graphene-based nanohybrids have been provided with recent advancements and opened up new possibilities to further improve their photodecomposition activity. 2D- sulfides-graphene-based photocatalysts can interact with different organic moieties and catalytic sulphur atoms, further improving their photodecomposition activity. In contrast, graphene attached with 2D-layer sulfides enhances electron mobility and efficient charge separations. Although sulfides-graphene-based nanohybrids have great potential owing to their fascinating properties, toxicity toward the environment and cost-effectiveness are the significant challenges we need to overcome to develop 2D-sulfides-graphene-based photocatalysts for industrial applications further.

Organic Molecules-Based Graphene Nanocomposites

Combining graphene or its derivatives with a specific class of organic molecules provides enhanced selectivity for the graphene layers (Zhang et al. 2020a, b, c, d; Su et al. 2023). Improved interaction with the targeted pollutant can occur

with the functionalization of these molecules with graphene. This unusual molecule-dependent selectivity makes this composite fascinating and essential in the field of environmental remediation and water detoxification. However, few reports are available on organic molecules-based graphene, which we have discussed below.

Organic molecules, natural and chemical, such as porphyrins, cyanine dyes, polyaniline, etc., have also been regarded as ambient light harvesters due to their sensitivity to UV light. These compounds have been associated with numerous photochemical reactions and electronic charge transfer processes. (Zhu et al. 2012b, 2013). These can thus be easily complexed with graphene to produce an efficient photocatalyst. Typically, the photocatalytic pathway followed by an organic graphene photocatalyst could be: a) Organic light harvester interacts with light to form excited species (A^*), which then transfers excess energy in the form of a hot electron to graphene, which further reacts with the analyte, thus triggering its degradation. At the same time, A^{++} accepts electrons from the donor molecules and returns to the ground state, maintaining the overall reaction dynamics. Light absorption by the organic molecule here is a function of the energy gap between the highest occupied and lowest unoccupied molecular orbital. Alternatively, b) organic photocatalyst can imitate a semiconductor photocatalyst system to produce charge carriers upon excitation, following a degradation pathway as explained in the previous section, where the photocarriers migrate to graphene and/or analyte on the surface to drive redox reactions, eventually leading to toxin disintegration. Larowska et al. (2020) reported the fabrication of porphyrins-graphene oxide nanocomposites using wet chemical methods and employed them to decompose RhB dye molecules under visible light. They prepared two nanocomposites, TMPyP-GO and ZnTMPyP-GO, with noncovalent attachment and explored the optical theory and charge transfer properties using several spectroscopic techniques and pinned down the effective charge transfer among graphene and porphyrins-based compounds. They have found that ZnTMPyP-GO revealed a higher performance as a photocatalyst due to the presence of Zn in the core of the porphyrins. In the photocatalytic test, they showed that using 0.4 mg photocatalyst sample for the decomposition of 5 mg/L solution of RhB in 120 min of visible light exposure. In another study, Ahadi et al. (2020) prepared manganese porphyrins/graphene oxide nanohybrids using an epoxidation process and embraced them for photocatalytic application. In their photocatalytic reaction, 0.50 mg of the synthesized Mn-porphyrins/graphene oxide nanohybrids were used for the photocatalytic conversion of the organic solution under LED light. They have demonstrated that with immobilization properties of Mn-porphyrin, significant enhancements in rate and enantioselectivity were achieved. Mitra et al. (2019) fabricated the polyaniline/RGO nanocomposite

using a polymerization chemical process and optimized the content of each constituent for an enhanced photodecomposition process. Their photocatalytic studies decomposed the different organic textile waste, namely MG, RhB, CR, and Cr ions solutions, under visible light illumination (200 W).

From the above discussion, we can conclude that Organic molecules-based Graphene photocatalysts contain essential properties such as multifunctionality, biocompatibility, and tunable optical properties, which are highly beneficial for boosting the photodecomposition ability significantly. Compared with the other graphene-based photocatalyst designs, this design has less environmental toxicity, is highly selective, multifunctional, and relatively inexpensive. For the development of organic molecules, the graphene nanohybrid photocatalyst suffers from the stability and scalability of commercial usage. Moreover, complex synthesis processes and optimization are significant hurdles that must be addressed for better performance in photocatalytic processes.

In our opinion, graphene layers with optimum defect concentration should be used to develop a highly efficient graphene-based photocatalyst. The other semiconductor should also contain superior optical properties and enhanced effective surface area. The proposed photocatalyst design should be hierarchical or layered structures that can interact well with the various organic pollutants. Apart from this, the band alignment of the semiconductor should provide effective charge separation, and their band position can produce unsaturated radicals, which can drive the photocatalysis process rapidly. Let's compare the design mentioned above of the proposed photocatalyst semiconductor-graphene-based. Photocatalyst provides the unique advantages of cost-effectiveness, stability, and efficiency compared to the other two designs. Although semiconductor-graphene composites require several improvements, there are a few challenges to developing these materials. On the other hand, for diverse practical and industrial applications, semiconductor-graphene composites are considered a superior candidate.

Challenges and Future Direction

As we have highlighted, different graphene-based nanocomposites reveal the remarkable photocatalytic efficiency of water detoxification applications. However, one of the primary challenges is optimizing the defect states with optimal functionalization while synthesizing graphene-based nanocomposite materials. Secondly, even after the different efforts of several research groups, large-scale production with the excellent performance of graphene-based nanocomposite photocatalyst has not been achieved. Another crucial issue for graphene-based nanocomposites includes the chemical stability, reusability, and assurances of the interfacial charge transfer among the graphene and another

Fig. 14 Challenges and probable solutions for the development of graphene-based photocatalyst

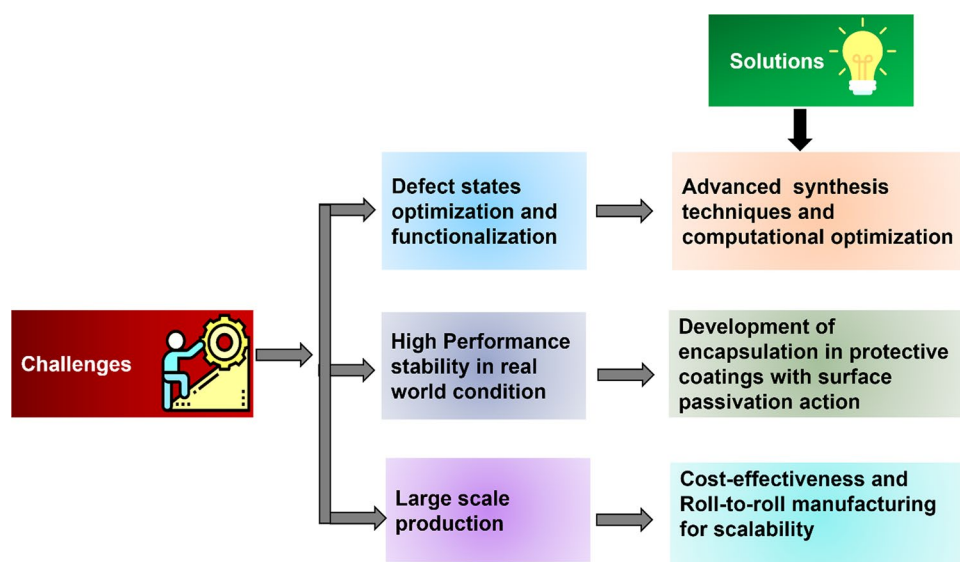
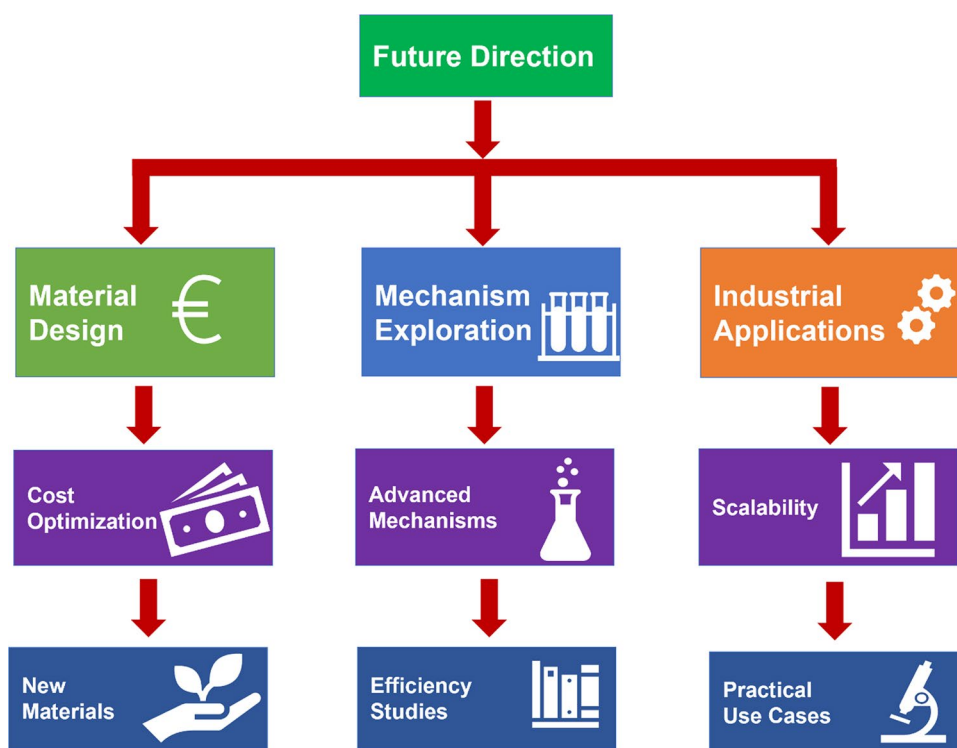


Fig. 15 Future direction for graphene-based photocatalyst for wastewater treatment



photocatalyst. Under real-world conditions, different light exposure, and real-time practical conditions, the stability of graphene can be varied, which can strongly influence its reusability, recyclability, and synergistic effect among the graphene and another photocatalyst sample (Kauling et al. 2018). The above-mentioned challenges and probable solutions have been illustrated in Fig. 14.

In our opinion, the future direction will emphasize improving the preparation method to provide the large-scale production of graphene-based photocatalysts

cost-effectively. Apart from this, biomaterials-derived synthesis should be developed for a sustainable environment. The advanced synthesis techniques and photocatalyst design should be formulated for precise tuning of different optical, electronic, and structural properties, which can provide the sufficient control over the functionalization and defect state optimization of graphene-based photocatalyst. Apart from this, the multi-component-based systems such as plasmonic-semiconductor-graphene, organic molecule-semiconductor-graphene, and plasmonic-organic molecule-graphene,

plasmonic-2D-sulfide-graphene-based nanohybrids photocatalyst should be developed which can provide the multifunctionality with enhanced photodecomposition activity due to efficient synergistic effect, improved optical properties and higher adsorption ability in a single platform. Advanced characterization techniques should be combined with the theoretical approach to provide an in-depth analysis of interfacial synergistic effects and charge carrier dynamics. The overview of the potential challenges and future directions is depicted in Fig. 15. Moreover, the performance abilities and different characterization of graphene-based photocatalysts should be designed for real-time conditions, which will be beneficial in developing the bridge between laboratory research and industrial-scale water detoxification solutions. Regarding different contaminations, advanced graphene-based nanohybrid photocatalyst-mediated redox reactions are being applied to treat bacteria-contaminated water and other applications such as H₂ production, CO₂ capture, and N₂ fixation.

Summary

In brief, using different photocatalyst materials, solar energy can be harvested and transferred into chemical energy for various environmental and energy production applications. The current review provides a basic overview of engineered graphene-based photocatalysts and their employment for water purification with the fundamental aspects and enriches discussion for understanding. As explained above, 2D-layered graphene-based nanostructures are fascinating due to their effective surface area and optical and electronic properties, which attract researchers to tailor their properties. Pure graphene with a metallic nature only harvests the UV region (~ 260 nm). In contrast, graphene oxide reveals a semiconductor nature, which provides flexibility in opting for the desired graphene-based compound per the required application. As explained above, graphene was initially employed as the co-catalyst. After further exploration, different composites with graphene-based compounds indicate outstanding activity for decomposing various organic pollutants under different light exposures. We have provided an overview of the graphene-based nanocomposite with different nanostructures such as plasmonic nanostructures, semiconductor metal oxides, 2D-layered nanostructures, and various organic molecules. This review highlights the primary charge transfer mechanism for various graphene-based compounds with other nanostructures, which develops a basic understanding of how readers can develop different 2D-layered materials for environmental applications. Moreover, we emphasize the properties of graphene and its nanohybrids and elucidate the correlation between these properties and their photocatalytic decomposition activity in

detail. We have also provided a clear comparison among the different existing designs of the graphene-based nanohybrid photocatalyst. A clear roadmap has been provided to enrich the discussion further, including probable challenges and future direction for developing graphene-based nanohybrid photocatalysts. This entails investigating novel preparation techniques, improving scalability, maximizing performance in realistic environmental settings, and creating multicomponent systems for combined applications in the energy and environmental domains.

Author Contributions Jaspal Singh: methodology, conceptualization data curation, investigation, visualization, resources, writing—original draft. D. Duc Nguyen: supervision, investigation, data curation, resources, writing—review and editing. Philippe Leclerc: conceptualization, resources, visualization, writing—review and editing. Phuong Nguyen-Tri: conceptualization, methodology, data curation, resources, investigation, visualization, writing—original draft—review and editing.

Data Availability The data presented in this study are available in the published research.

Declarations

Conflict of interest The authors declare no conflict of interest.

References

- Aba Z, Goktas A, Kilic A (2024) Characterization of Zn_{1-x}La_xS thin films; compositional, surface, optical, and photoluminescence properties for possible optoelectronic and photocatalytic applications. *J Sol-Gel Sci Technol* 109:260–271
- Abid, Sehrawat, P, Islam SS, Mishra P, Ahmad S (2018) Reduced graphene oxide (rGO)-based wideband optical sensor and the role of temperature, defect states, and quantum efficiency. *Sci Rep* 8:3537
- Adán-Más A, Wei D (2013) Photoelectrochemical properties of graphene and its derivatives. *Nanomaterials* 3:325–356
- Ahadi E, Hosseini-Monfared H, Spieb A, Janiak C (2020) Photocatalytic asymmetric epoxidation of trans-stilbene with manganese-porphyrin/graphene-oxide nanocomposite and molecular oxygen: axial ligand effect. *Catal Sci Technol* 10:3290–3302
- Ali J, Guo S, Chen Y, Shahzad A, Ullah MW, Chen F (2024) Metal sulfides as emerging materials for advanced oxidation of wastewater: recent developments, challenges, and prospects. *Coordination Chem Rev* 509:215765
- An X, Yu CJ, Wang F, Li C, Li Y (2013) One-pot synthesis of In₂S₃ nanosheets/graphene composites with enhanced visible-light photocatalytic activity. *Appl Catal B* 129:80–88
- Anichini C, Samori P (2021) Graphene-based hybrid functional materials. *Small* 17:2100514
- Aroob S, Taj MB, Shabbir S, Imran M, Ahmad RH, Habib S, Raheel A, Akhtar MN, Ashfaq M, Sillanpää M (2021) In situ biogenic synthesis of CuO nanoparticles over graphene oxide: a potential nanohybrid for water treatment. *J Environ Chem Eng* 9:105590
- Arshad A, Iqbal J, Mansoor Q (2017) NiO-nanoflakes grafted graphene: an excellent photocatalyst and a novel nanomaterial for achieving complete pathogen control. *Nanoscale* 9:16321–16328

- Aslan E, Sahin G, Goktas A (2023) Facile synthesis of Sb_2S_3 micro-materials for highly sensitive visible light photodetectors and photocatalytic applications. *Mater Chem Phys* 307:128160
- Aslan E, Emir Ö, Arslan F, Goktas A, Tumbul A, Durgun M, Kilic A, Aktacir MA, Aslan F (2024) Improving the optical properties of CuCoMnO_x spinel absorber using ZnO nanorod arrays for thermal collector and photocatalytic applications. *Ceramics Int* 50:9169–9176
- Avouris P (2010) Graphene: electronic and photonic properties and devices. *Nano Lett* 10:4285–4294
- Bhunia SK, Jana NR (2014) Reduced graphene oxide-silver nanoparticle composite as visible light photocatalyst for degradation of colorless endocrine disruptors. *ACS Appl Mater Interfaces* 6:20085–20092
- Bhushan R, Kumar P, Thakur AK (2020) Catalyst-free solvothermal synthesis of ultrapure elemental N- and B-doped graphene for energy storage application. *Solid State Ionics* 353:115371
- Biroju RK, Choudhury B, Giri PK (2016) Plasmon-enhanced strong visible light photocatalysis by defect engineered CVD graphene and graphene oxide physically functionalized with Au nanoparticles. *Catal Sci Technol* 6:7101–7112
- Borane N, Boddula R, Odedara N, Singh J, Andhe M, Patel R (2024) Comprehensive review on synthetic methods and functionalization of graphene oxide: emerging applications. *Nano-Struct Nano-Objects* 39:101282
- Burkholder MB, Rahman FBA, Chandler EH Jr, Regalbuto JR, Gupton BF, Tengco JMM (2022) Metal supported graphene catalysis: a review on the benefits of nanoparticulate supported specialty sp^2 carbon catalysts on enhancing the activities of multiple chemical transformations. *Carbon Trends* 9:100196
- Busarello P, de Quadros S, Zimmermann LM, Neiva EGC (2023) Graphene oxide/ ZnO nanocomposites applied in photocatalysis of dyes: tailoring aqueous stability of quantum dots. *Colloids Surf A* 675:132026
- Chai B, Li J, Xu Q (2014) Reduced graphene oxide grafted Ag_3PO_4 composites with efficient photocatalytic activity under visible-light irradiation. *Indus Eng Chem Res* 53:8744–8752
- Chakraborty S, Mukherjee A, Basu S (2018) RGO- MoS_2 supported NiCo_2O_4 catalyst toward solar water splitting and dye degradation. *ACS Sus Chem Eng* 6:5238–5247
- Chan SHS, Wu TY, Juan JC, The CY (2011) Recent developments of metal oxide semiconductors as photocatalysts in advanced oxidation processes (AOPs) for treatment of dye wastewater. *J Chem Technol Biotechnol* 86:1130–1158
- Chauhan H, Soni K, Kumar M, Deka S (2016) Tandem photocatalysis of graphene-stacked SnS_2 nanodiscs and nanosheets with efficient carrier separation. *ACS Omega* 1:127–137
- Chen D, Lin Z, Sartin MM, Huang TX, Liu J, Zhang Q, Han L, Li JF, Tian ZQ, Zhan D (2020a) Photosynergetic electrochemical synthesis of graphene oxide. *J Am Chem Soc* 142:6516–6520
- Chen LH, Shen HT, Chang WH, Khalil I, Liao SY, Yehye WA, Liu SC, Chu CC, Hsiao VKS (2020b) Photocatalytic properties of graphene/gold and graphene oxide/gold nanocomposites synthesized by pulsed laser induced photolysis. *Nanomaterials* 10:1985
- Cho KM, Kim KH, Choi HO, Jung HT (2015) A highly photoactive, visible-light-driven graphene/2D mesoporous TiO_2 photocatalyst. *Green Chem* 17:3972–3978
- Cousins IT, Johansson JH, Salter ME, Sha B, Scherlinger M (2022) Outside the safe operating space of a new planetary boundary for per- and polyfluoroalkyl substances (PFAS). *Environ Sci Technol* 56:11172–11179
- Darabdhara G, Boruah PK, Borthakur P, Hussain N, Das MR, Ahamad T, Alshehri SM, Malgras V, Wu KCW, Yamauchi Y (2016) Reduced graphene oxide nanosheets decorated with Au-Pd bimetallic alloy nanoparticles towards efficient photocatalytic degradation of phenolic compounds in water. *Nanoscale* 8:8276–8287
- Dashairya L, Sharma M, Basu S, Saha P (2019) SnS_2/RGO based nanocomposite for efficient photocatalytic degradation of toxic industrial dyes under visible-light irradiation. *J Alloys Compds* 774:625–636
- Di Bartolomeo A (2016) Graphene Schottky diodes: an experimental review of the rectifying graphene/semiconductor heterojunction. *Phys Rep* 606:1–58
- Dodds JN, Alexander NLM, Kirkwood KI, Foster MR, Hopkins ZR, Knappe DRU, Baker ES (2020) From pesticides to per- and polyfluoroalkyl substances: an evaluation of recent targeted and untargeted mass spectrometry methods for xenobiotics. *Anal Chem* 93:641–656
- Dridi D, Vu NN, Singh J, Eesaee M, Saidi A, Elkoun S, Nguyen-Tri P (2024) Recent advances on engineering of silver-related nanocomposites toward antibacterial applications. *Nano-Struct Nano-Objects* 38(101):195
- Dubey AK, Yadav RK, Gole VL, Sharma K, Singh S, Shahin R, Mishra S, Baeg JO, Gupta NK (2024) Functionalized nitrogen-enriched graphene as a highly efficient photocatalyst for oxidative cyclization of thioamide and degradation of dye. *J Mol Stru* 1312:138557
- Fang B, Chang D, Xu Z, Gao C (2020) A review on graphene fibers: expectations, advances, and prospects. *Adv Mater* 32:1902664
- Felis E, Buta-Hubeny M, Zieliński W, Hubeny J, Harnisz M, Bajkacz S, Korzeniewska E (2022) Solar-light driven photodegradation of antimicrobials, their transformation by-products, and antibiotic resistance determinants in treated wastewater. *Sci Total Environ* 36:155447
- Feng H, Cheng R, Zhao X, Duan X, Li J (2013) A low-temperature method to produce highly reduced graphene oxide. *Nature Comm* 4:1539
- Fujishima A, Honda K (1972) Electrochemical photolysis of water at a semiconductor electrode. *Nature* 238:37–38
- Gallerati A (2022) Graphene, Dirac equation and analogue gravity. *Physical Scri* 97:064005
- Gao D, Liu X, Junaid M, Liao H, Chen G, Wu Y, Wang J (2022) Toxicological impacts of micro(nano)plastics in the benthic environment. *Sci Total Environ* 836:155620
- Garg S, Chandra A (2022) Green photocatalytic semiconductors. Springer, New York
- Gayathri S, Jayabal P, Kottaisamy M, Ramakrishnan V (2014) Synthesis of ZnO decorated graphene nanocomposite for enhanced photocatalytic properties. *J Appl Phys* 115:173504
- Georgakilas V, Otyepka M, Bourlinos AB, Chandra V, Kim N, Kemp KC, Hobza P, Zboril R, Kim KS (2012) Functionalization of graphene: covalent and non-covalent approaches, derivatives and applications. *Chem Rev* 112:6156–6214
- Ghasemi S, Hashemian SJ, Alamolhoda AA, Gocheva I, Rahman Setayesh S (2017) Plasmon enhanced photocatalytic activity of $\text{Au}@\text{TiO}_2$ -graphene nanocomposite under visible light for degradation of pollutants. *Mater Res Bull* 87:40–47
- Goktas S, Goktas A (2021) A comparative study on recent progress in efficient ZnO -based nanocomposite and heterojunction photocatalysts: a review. *J Alloys Compds* 863:158734
- Goktas A, Aslan E, Arslan F, Kilic A (2022a) Characterization of multifunctional solution-processed $\text{Sn}_{1-x}\text{Zn}_x\text{S}$ nanostructured thin films for photosensitivity and photocatalytic applications. *Opt Mater* 133:112984
- Goktas A, Modanlı S, Tumbul A, Kilic A (2022b) Facile synthesis and characterization of ZnO , $\text{ZnO}:\text{Co}$, and $\text{ZnO}/\text{ZnO}:\text{Co}$ nanorod-like homojunction thin films: role of crystallite/grain size and microstrain in photocatalytic performance. *J Alloys Compds* 893:162334

- Göktaş S (2024) Synergic effects of pH, reaction temperature, and various light sources on the photodegradation of methylene blue without photocatalyst: a relatively high degradation efficiency. *Chem Afr* 7:4425–4437
- Göktaş S, Tumbul A, Göktaş A (2023) Growth technique-induced highly C-axis-oriented ZnO: Mn, ZnO: Fe, and ZnO: Co thin films: a comparison of nanostructure, surface morphology, optical band gap, and room-temperature ferromagnetism. *J Supercond Novel Magn* 36:1875–1892
- Guirguis A, Polaki SR, Sahoo G, Ghosh S, Kamruddin M, Merenda A, Chen X, Maina JW, Szekely G, Dumee L (2020) Engineering high-defect densities across vertically-aligned graphene nanosheets to induce photocatalytic reactivity. *Carbon* 168:32–41
- Guo S, Garaj S, Bianco A, Ménard-Moyon C (2022) Controlling covalent chemistry on graphene oxide. *Nat Rev Phys* 4:247–262
- Han C, Zhang N, Xu YJ (2016) Structural diversity of graphene materials and their multifarious roles in heterogeneous photocatalysis. *Nano Today* 11:351–372
- Han C, Li YH, Qi MY, Zhang F, Tang ZR, Xu YJ (2020) Surface/interface engineering of carbon-based materials for constructing multidimensional functional hybrids. *Solar RRL* 4:1900577
- Hong X, Wang X, Li Y, Fu J, Liang B (2020) Progress in graphene/metal oxide composite photocatalysts for degradation of organic pollutants. *Catalysts* 10:921
- Hsiao MC, Liao SH, Yen MY, Liu PI, Pu NW, Wang CA, Ma CCM (2010) Preparation of covalently functionalized graphene using residual oxygen-containing functional groups. *ACS Appl Mater Interfaces* 2:3092–3099
- Hu X, Meng X, Zhang Z (2016) Synthesis and characterization of graphene oxide-modified Bi₂WO₆ and its use as photocatalyst. *Int J Photoenergy* 2016:8730806
- Huang Z, Wang J, Lu S, Xue H, Chen Q, Yang MQ, Qian Q (2021) Insight into the real efficacy of graphene for enhancing photocatalytic efficiency: a case study on CVD graphene-TiO₂ composites. *ACS Appl Energy Mater* 4:8755–8764
- Ibrahim A, Klopocinska A, Horvat K, Abdel Hamid Z (2021) Graphene-based nanocomposites: synthesis, mechanical properties, and characterizations. *Polymers* 13:2869
- Ji L, Meduri P, Agubra V, Xiao X, Alcoutlabi M (2016) Graphene-based nanocomposites for energy storage. *Adv Energy Mater* 6:1502159
- Jiang Y, Chowdhury S, Balasubramanian R (2019) Nitrogen and sulfur codoped graphene aerogels as absorbents and visible light-active photocatalysts for environmental remediation applications. *Environ Poll* 251:344–353
- Jiang B, Chen D, Li N, Xu Q, Li H, Lu J (2023) Graphene-encapsulated CdS as a high-performance photocatalyst for H₂O₂ production from pure water. *Indus Eng Chem Res* 62:12974–12984
- Kauling AP, Seefeldt AT, Pisoni DP, Pradeep RC, Bentini R, Oliveira RVB, Novoselov KS, Castro Neto AH (2018) The worldwide graphene flake production. *Adv Mater* 30:1803784
- Kemp KC, Chandra V, Saleh M, Kim KS (2013) Reversible CO₂ adsorption by an activated nitrogen-doped graphene/polyaniline material. *Nanotechnology* 24:235703
- Khan M, Assal ME, Tahir MN, Khan M, Ashraf M, Hatshan MR, Khan M, Varala R, Badawi NM, Adil SF (2022) Graphene/inorganic nanocomposites: evolving photocatalysts for solar energy conversion for environmental remediation. *J Saudi Chem Soc* 26:101544
- Khin MM, Nair AS, Babu VJ, Murugan R, Ramakrishna S (2012) A review on nanomaterials for environmental remediation. *Energy Environ Sci* 5:8075–8109
- Kuang P, Sayed M, Fan J, Cheng B, Yu J (2020) 3D graphene-based H₂-production photocatalyst and electrocatalyst. *Adv Energy Mater* 10:1903802
- Kumar A, Choudhary P, Kumar A, Camargo PHC, Krishnan V (2022) Recent advances in plasmonic photocatalysis based on TiO₂ and noble metal nanoparticles for energy conversion, environmental remediation, and organic synthesis. *Small* 18:2101638
- Kumar ED, Easwaramoorthi S, Rao JR (2023) Gold-reduced graphene oxide intimate BiVO₄-ZnO mixed oxide composite with leveraged charge carrier transport under solar radiation. *Opt Mater* 142:114054
- Kwon KC, Choi KS, Kim SY (2012) Increased work function in few-layer graphene sheets via metal chloride doping. *Adv Fun Mater* 22:4724–4731
- Larowska D, O'Brien JM, Senge MO, Burdzinski G, Marciniak B, Lewandowska-Andralojc A (2020) Graphene oxide functionalized with cationic porphyrins as materials for the photodegradation of rhodamine B. *J Phys Chem C* 124:15769–15780
- Lee E, Hong JY, Kang H, Jang J (2012) Synthesis of TiO₂ nanorod-decorated graphene sheets and their highly efficient photocatalytic activities under visible-light irradiation. *J Hazard Mater* 219:13–18
- Li X, Yu J, Wageh S, Al-Ghamdi AA, Xie J (2016) Graphene in photocatalysis: a review. *Small* 12:6640–6696
- Li B, Zhang S, Cui C, Qian W, Jin Y (2022a) Comprehensive review on nitrogen-doped graphene: Structure characterization, growth strategy, and capacitive energy storage. *Energy Fuels* 37:902–918
- Li J, Chen M, Samad A, Dong H, Ray A, Zhang J, Jiang X (2022b) Wafer-scale single-crystal monolayer graphene grown on sapphire substrate. *Nat Mater* 21:740–747
- Li L, Zhang R, Hou P, Lin Y, Wang D, Xie T (2023) Boosting hydrogenation of graphene quantum dot-modified photocatalysts: specific functionalized modulation at active sites. *ACS Catal* 13:10824–10834
- Liu M, Xue X, Yu S, Wang X, Hu X, Tian H, Chen H, Zheng W (2017) Improving photocatalytic performance from Bi₂WO₆@MoS₂/graphene hybrids via gradual charge transferred pathway. *Sci Rep* 7:3637
- Liu G, Han K, Zhou Y, Ye H, Zhang X, Hu J, Li X (2018) Facile synthesis of highly dispersed Ag doped graphene oxide/titanate nanotubes as a visible light photocatalytic membrane for water treatment. *ACS Sus Chem Eng* 6:6256–6263
- Liu K, Chen J, Sun F, Liu Y, Tang M, Yang Y (2022) Historical development and prospect of intimately coupling photocatalysis and biological technology for pollutant treatment in sewage: a review. *Sci Total Environ* 835:155482
- Liu X, Yang X, Cui J, Wu C, Sun Y, Du X, Chen J, Ye J, Liu L (2023) Ni coated with N-doped graphene layer as active and stable H₂ evolution cocatalysts for photocatalytic overall water splitting. *ACS Catal* 13:14314–14323
- Liu YL, Li D, Cao P, Yin X, Zeng Q, Zhou H (2024) Advances in MXene-based composite materials for efficient removal of radioactive nuclides and heavy metal ions. *Mater Today Phys* 44:101444
- Lu G, Yu K, Wen Z, Chen J (2013) Semiconducting graphene: converting graphene from semimetal to semiconductor. *Nanoscale* 5:1353–1368
- Lu KQ, Zhang N, Han C, Li F, Chen Z, Xu YJ (2016) Insight into the origin of boosted photosensitive efficiency of graphene from the cooperative experiment and theory study. *J Phys Chem C* 120:27091–27103
- Lu KQ, Xin X, Zhang N, Tang ZR, Xu YJ (2018) Photoredox catalysis over graphene aerogel-supported composites. *J Mater Chem A* 6:4590–4604
- Lu KQ, Li YH, Tang ZR, Xu YJ (2021) Roles of graphene oxide in heterogeneous photocatalysis. *ACS Mater Au* 1:37–54
- Luo Y, Li M, Hu G, Tang T, Wen J, Li X, Wang L (2018) Enhanced photocatalytic activity of sulfur-doped graphene quantum

- dots decorated with TiO₂ nanocomposites. *Mater Res Bull* 97:428–435
- Lyu H, Li P, Tang J, Zou W, Wang P, Gao B, Dong L (2023) Single-atom Mn anchored on N-doped graphene oxide for efficient adsorption-photocatalytic degradation of sulfanilamide in water: electronic interaction and mineralization pathway. *Chem Eng J* 454:140120
- Maarissetty D, Baral SS (2020) Defect engineering in photocatalysis: formation, chemistry, optoelectronics, and interface studies. *J Mater Chem A* 8:18560–18604
- Manchala S, Nagappagari LR, Venkatakrishnan SM, Shanker V (2019) Solar-light harvesting bimetallic Ag/Au decorated graphene plasmonic system with efficient photoelectrochemical performance for the enhanced water reduction process. *ACS Appl Nano Mater* 2:4782–4792
- Manzeli S, Ovchinnikov D, Pasquier D, Yazyev OV, Kis A (2017) 2D transition metal dichalcogenides. *Nat Rev Mater* 2:1–15
- Merino-Díez N, García-Lekue A, Carbonell-Sanromá E, Li J, Corso M, Colazzo L, Sedona F, Sánchez-Portal D, Pascual JI, de Oteyza DG (2017) Width-dependent band gap in armchair graphene nanoribbons reveals Fermi level pinning on Au (111). *ACS Nano* 11:11661–11668
- Min S, Lu G (2011) Dye-sensitized reduced graphene oxide photocatalysts for highly efficient visible-light-driven water reduction. *J Phys Chem C* 115:13938–13945
- Minella M, Sordello F, Minero C (2017) Photocatalytic process in TiO₂/graphene hybrid materials. evidence of charge separation by electron transfer from reduced graphene oxide to TiO₂. *Catal Today* 281:29–37
- Mitra M, Ahamed ST, Ghosh A, Mondal A, Kargupta K, Ganguly S, Banerjee D (2019) Polyaniline/reduced graphene oxide composite-enhanced visible-light-driven photocatalytic activity for the degradation of organic dyes. *ACS Omega* 4:1623–1635
- Mohaghegh N, Tasviri M, Rahimi E, Gholami MR (2015) Comparative studies on Ag₃PO₄/BiPO₄-metal-organic framework-graphene-based nanocomposites for photocatalysis application. *Appl Surf Sci* 351:216–224
- Moon IK, Lee J, Ruoff RS, Lee H (2010) Reduced graphene oxide by chemical graphitization. *Nat Comm* 1:73
- Naldoni A, Altomare M, Zoppellaro G, Liu N, Kment S, Zboril R, Schmuki P (2018) Photocatalysis with reduced TiO₂: From black TiO₂ to cocatalyst-free hydrogen production. *ACS Catal* 9:345–364
- Nasr M, Benhamou L, Kotbi A, Rajput NS, Campos A, Lahmar AI, Hoummada K, Kaja K, El Marssi M, Jouiad M (2022) Photoelectrochemical enhancement of graphene@WS₂ nanosheets for water splitting reaction. *Nanomaterials* 12:1914
- Nebol'sin VA, Galstyan V, Silina YE (2020) Graphene oxide and its chemical nature: multi-stage interactions between the oxygen and graphene. *Surf Interfaces* 21:100763
- Nemati F, Rezaie M, Tabesh H, Eid K, Xu G, Ganjali MR, Hosseini M (2022) Cerium functionalized graphene nano-structures and their applications: a review. *Environ Res* 208:112685
- Nugraheni AY, Nasrullah M, Prasetya FA, Astuti F, Darminto, (2015) Study on phase, molecular bonding, and bandgap of reduced graphene oxide prepared by heating coconut shell. *Mater Sci Forum* 827:285–289
- Padmanabhan NT, Thomas N, Louis J, Mathew DT, Ganguly P, John H, Pillai SC (2021) Graphene coupled TiO₂ photocatalysts for environmental applications: a review. *Chemosphere* 271:129506
- Papageorgiou DG, Kinloch IA, Young RJ (2017) Mechanical properties of graphene and graphene-based nanocomposites. *Prog Mater Sci* 90:75–127
- Parrino F, Livraghi S, Giamello E, Ceccato R, Palmisano L (2020) Role of hydroxyl, superoxide, and nitrate radicals on the fate of bromide ions in photocatalytic TiO₂ suspensions. *ACS Catal* 10:7922–7931
- Pastrana-Martínez LM, Morales-Torres S, Likodimos V, Falaras P, Figueiredo JL, Faria JL, Silva AMT (2014) Role of oxygen functionalities on the synthesis of photocatalytically active graphene-TiO₂ composites. *Appl Catal B* 158:329–340
- Pei L, Yuan Y, Bai W, Li T, Zhu H, Ma Z, Zhong J, Yan S, Zou Z (2020) In situ-grown island-shaped hollow graphene on TaON with spatially separated active sites achieving enhanced visible-light CO₂ reduction. *ACS Catal* 10:15083–15091
- Pervez MN, Wei Y, Sun P, Qu G, Naddeo V, Zhao Y (2021) α -FeOOH quantum dots impregnated graphene oxide hybrids enhanced arsenic adsorption: the mediation role of environmental organic ligands. *Sci Total Environ* 781:146726
- Pham PV, Bodepudi SC, Shehzad K, Liu Y, Xu Y, Yu B, Duan X (2022) 2D heterostructures for ubiquitous electronics and optoelectronics: principles, opportunities, and challenges. *Chem Rev* 122:6514–6613
- Pogacean F, Socaci C, Pruneanu S, Biris AR, Coros M, Magerusan L, Katona G, Turcu R, Borodi G (2015) Graphene-based nanomaterials as chemical sensors for hydrogen peroxide-A comparison study of their intrinsic peroxidase catalytic behavior. *Sens Actuators B* 213:474–483
- Prakash J (2022) Mechanistic insights into graphene oxide driven photocatalysis as co-catalyst and sole catalyst in degradation of organic dye pollutants. *Photochemistry* 2:651–671
- Pramanik A, Thakur S, Singh B, Willke P, Wenderoth M, Hofsäss H, Di Santo G, Petaccia L, Maiti K (2022) Anomalies at the Dirac point in graphene and its hole-doped compositions. *Phys Rev Lett* 128:166401
- Quan Q, Lin X, Zhang N, Xu YJ (2017) Graphene and its derivatives as versatile templates for materials synthesis and functional applications. *Nanoscale* 9:2398–2416
- Radadiya TM (2015) A properties of graphene. *Euro J Mater Sci* 2:6–18
- Ravi A, Kumar MP, Rekha MY, Santosh MS, Srivastava C (2020) Graphene based nanocomposites: synthesis, properties and application as electrochemical sensors. *Comprehensive analytical chemistry*, vol 91. Elsevier, Amsterdam, pp 1–20
- Ren R, Li S, Li J, Ma J, Liu H, Ma J (2015) Enhanced catalytic activity of Au nanoparticles self-assembled on thiophenol functionalized graphene. *Catal Sci Technol* 5:2149–2156
- Rosman NN, Mohamad Yunus R, Minggu LJ, Arifin K, Salehmin MNI, Mohamed MA, Kassim MB (2018) Photocatalytic properties of two-dimensional graphene and layered transition-metal dichalcogenides based photocatalyst for photoelectrochemical hydrogen generation: an overview. *Int J Hydrogen Energy* 43:18925–18945
- Roy P, Periasamy AP, Liang CT, Chang HT (2013) Synthesis of graphene-ZnO-Au nanocomposites for efficient photocatalytic reduction of nitrobenzene. *Environ Sci Technol* 47:6688–6695
- Rybin M, Pereyaslavtsev A, Vasilieva T, Myasnikov V, Sokolov I, Pavlova A, Obratsova E, Khomich A, Ralchenko V, Obratsova E (2016) Efficient nitrogen doping of graphene by plasma treatment. *Carbon* 96:196–202
- Sahin G, Goktas A, Aslan E (2024) A new way to tune photocatalytic activity, surface morphology, and structural/optical parameters of ZrO₂ thin films using different Zr sources along with annealing temperature and film thickness. *J Sol-Gel Sci Technol* 112:425–443
- Sawant SY, Cho MH (2015) Facile electrochemical assisted synthesis of ZnO/graphene nanosheets with enhanced photocatalytic activity. *RSC Adv* 5:97788–97797
- Schweitzer L, Noblet J (2018) Water contamination and pollution. In: Trk B, Dransfield T (eds) *Green Chemistry*. Elsevier, Amsterdam, pp 261–290

- Shafiee A, Iravani S, Varma RS (2022) Graphene and graphene oxide with anticancer applications: challenges and future perspectives. *MedComm* 3:118
- Shah NRAM, Mohamad Yunus R, Rosman NN, Wong WY, Arifin K, Minggu LJ (2021) Current progress on 3D graphene-based photocatalysts: from synthesis to photocatalytic hydrogen production. *Int J Hydrogen Energy* 46:9324–9340
- Shah SJ, Luan X, Yu X, Su W, Wang Y, Zhao Z, Zhao Z (2024) Construction of 3D-graphene/NH₂-MIL-125 nanohybrids via amino-ionic liquid dual-mode bonding for advanced acetaldehyde photodegradation under high humidity. *J Colloids Interface Sci* 663:491–507
- Shanmugam M, Alsalmeh A, Alghamdi A, Jayavel R (2015) Enhanced photocatalytic performance of the graphene-V₂O₅ nanocomposite in the degradation of methylene blue dye under direct sunlight. *ACS Appl Mater Interfaces* 7:14905–14911
- Sharma R, Almasi M, Nehra SP, Rao VS, Panchal P, Rattan Paul D, Jain IP, Sharma A (2022) Photocatalytic hydrogen production using graphitic carbon nitride (GCN): a precise review. *Renew Sustain Energy Rev* 168:112776
- Shi Z, Ci H, Yang X, Liu Z, Sun J (2022) Direct-chemical vapor deposition-enabled graphene for emerging energy storage: versatility, essentiality, and possibility. *ACS Nano* 16:11646–11675
- Shiravizadeh AG, Yousefi R, Elahi SM, Sebt SA (2017) Effects of annealing atmosphere and rGO concentration on the optical properties and enhanced photocatalytic performance of SnSe/rGO nanocomposites. *Phys Chem Chem Phys* 19:18089–18098
- Shown I, Hsu HC, Chang YC, Lin CH, Roy PK, Ganguly A, Wang CH (2014) Highly efficient visible light photocatalytic reduction of CO₂ to hydrocarbon fuels by Cu-nanoparticle decorated graphene oxide. *Nano Lett* 14:6097–6103
- Singh J, Soni RK (2020a) Controlled synthesis of CuO decorated defect-enriched ZnO nanoflakes for improved sunlight-induced photocatalytic degradation of organic pollutants. *Appl Surf Sci* 521:146420
- Singh J, Soni RK (2020b) Fabrication of hydroxyl group-enriched mixed-phase TiO₂ nanoflowers consisting of nanoflakes for efficient photocatalytic activity. *J Mater Sci Mater Electron* 31:12546–12560
- Singh J, Soni RK (2020c) Two-dimensional MoS₂ nanosheet-modified oxygen defect-rich TiO₂ nanoparticles for light emission and photocatalytic applications. *New J Chem* 44:14936–14946
- Singh J, Soni RK (2021a) Enhanced sunlight-driven photocatalytic activity of In₂S₃ nanosheets functionalized MoS₃ nanoflowers heterostructure. *Sci Rep* 11:15352
- Singh J, Soni RK (2021b) Fabrication of nanostructured In₂S₃ thin film with broad optical absorption for improved sunlight-mediated photocatalysis application. *Opt Mater* 122:111748
- Singh J, Soni RK (2021c) Tunable optical properties of Au nanoparticles encapsulated TiO₂ spheres and their improved sunlight-mediated photocatalytic activity. *Colloids Surf A* 612:126011
- Singh J, Soni RK (2021d) Efficient charge separation in Ag nanoparticles functionalized ZnO nanoflakes/CuO nanoflowers hybrids for improved photocatalytic and SERS activity. *Colloids Surf A* 626:127005
- Singh J, Singh R, Duhan P, Mann Y, Manna AK, Banerjee D, Soni RK (2021e) Morphology dependent effective charge separation process in nanostructured MoS₂ thin films for enhanced photodegradation behavior. *J Phys D: Appl Phys* 54:375103
- Singh J, Juneja S, Soni RK, Bhattacharya J (2021f) Sunlight mediated enhanced photocatalytic activity of TiO₂ nanoparticles functionalized CuO-Cu₂O nanorods for removal of methylene blue and oxytetracycline hydrochloride. *J Coll Interface Sci* 590:60–71
- Singh M, Kaushal S, Singh P, Sharma J (2018) Boron-doped graphene oxide with enhanced photocatalytic activity for organic pollutants. *J Photochem Photobiol A* 364:130–139
- Singh J, Juneja S, Palsaniya S, Manna AK, Soni RK, Bhattacharya J (2019a) Evidence of oxygen defects-mediated enhanced photocatalytic and antibacterial performance of ZnO nanorods. *Colloids Surf B* 184:110541
- Singh J, Manna AK, Soni RK (2019b) Bifunctional Au-TiO₂ thin films with enhanced photocatalytic activity and SERS based multiplexed detection of organic pollutant. *J Mater Sci Mater Electron* 30:16478–16493
- Singh J, Kumar S, Soni RK (2020a) Synthesis of 3D-MoS₂ nanoflowers with tunable surface area for application in photocatalysis and SERS-based sensing. *J Alloys Compds* 849:156502
- Singh J, Kumar S, Manna AK, Soni RK (2020b) Fabrication of ZnO-TiO₂ nanohybrids for rapid sunlight-driven photodegradation of textile dyes and antibiotic residue molecules. *Opt Mater* 107:110138
- Singh J, Kumar S, Verma HK, Soni RK (2020c) Cost-effective scalable synthesis of few layers MoS₂ based thin film for sunlight enforced photocatalytic activity. *Opt Mater* 110:110506
- Singh J, Manna AK, Soni RK (2020d) Sunlight-driven photocatalysis and non-enzymatic glucose sensing performance of cubic structured CuO thin films. *Appl Surf Sci* 530:147258
- Singh J, Juneja S, Soni RK, Bhattacharya J (2021a) Sunlight-mediated enhanced photocatalytic activity of TiO₂ nanoparticles functionalized CuO-Cu₂O nanorods for removal of methylene blue and oxytetracycline hydrochloride. *J Colloids Interface Sci* 590:60–71
- Singh J, Singh R, Duhan P, Mann Y, Manna AK, Banerjee D, Soni RK (2021b) Morphology-dependent effective charge separation process in nanostructured MoS₂ thin films for enhanced photodegradation behavior. *J Phys D* 54:375103
- Singh J, Akhtar S, Tran TT, Kim J (2023a) MoS₂ nanoflowers functionalized with C₃N₄ nanosheets for enhanced photodecomposition. *J Alloys Compds* 954:170206
- Singh J, Roy S, Tran TT, Akhtar S, Lee E, Kim J (2023b) Photocatalytic thin films based on Au nanoparticles covered by iron oxyhydroxides by hydrothermal process. *Surf Interfaces* 39:102915
- Singh J, Soni RK, Nguyen DD, Gupta VK, Nguyen-Tri P (2023c) Enhanced photocatalytic and SERS performance of Ag nanoparticles functionalized MoS₂ nanoflakes. *Chemosphere* 339:139735
- Singh J, Verma AK (2023) Plasmonic-TiO₂ nanohybrid for environmental and energy applications. In *Updates on Titanium Dioxide*. IntechOpen.
- Song SM, Park JK, Sul OJ, Cho BJ (2012) Determination of work function of graphene under a metal electrode and its role in contact resistance. *Nano Lett* 12:3887–3892
- Song Y, Massuyeau F, Jiang L, Dan Y, Le Rendu P, Nguyen TP (2019a) Effect of graphene size on the photocatalytic activity of TiO₂/poly (3-hexylthiophene)/graphene composite films. *Catal Today* 321:74–80
- Song Z, Ma YL, Li CE (2019b) The residual tetracycline in pharmaceutical wastewater was effectively removed by using MnO₂/graphene nanocomposite. *Sci Total Environ* 651:580–590
- Song Y, Gao Y, Liu X, Ma J, Chen B, Xie Q, Gao X (2022) Transfer-enabled fabrication of graphene wrinkle arrays for epitaxial growth of AlN films. *Adv Mater* 34:2105851
- Sreepasad TS, Berry V (2013) How do the electrical properties of graphene change with its functionalization? *Small* 9:341–350
- Su Y, Lin Y, Li Y, Ren T, Deng Y, Zheng C (2023) A high-throughput atomic emission analyzer for simultaneous field detection of dissolved inorganic and organic carbon in seawater and lake water. *Anal Chim Acta* 1261:341184
- Sui D, Xu L, Zhang H, Sun Z, Kan B, Ma Y, Chen Y (2020) A 3D cross-linked graphene-based honeycomb carbon composite with excellent confinement effect of organic cathode material for lithium-ion batteries. *Carbon* 157:656–662

- Tai XH, Lai CW, Yang TCK, Lee KM, Boonyuen S, Jiwanti PK, Juan JC (2024) Enrichment of graphitic-N in nitrogen-doped photoreduced graphene oxide as highly efficient photocatalyst for mineralization of volatile organic compounds. *Mater Today Chem* 42:102372
- Tu W, Zhou Y, Zou Z (2013) Versatile graphene-promoting photocatalytic performance of semiconductors: basic principles, synthesis, solar energy conversion, and environmental applications. *Adv Funct Mater* 23:4996–5008
- Tumbul A, Aslan E, Göktaş A, Mutlu IH, Arslan F, Aslan F (2024) Chemically derived quinary $\text{Cu}_2\text{Co}_{1-x}\text{Na}_x\text{SnS}_4$ photon absorber material and its photocatalytic application. *Appl Phys A* 130:225
- Vazirisereshk MR, Ye H, Ye Z, Otero-De-La-Roza A, Zhao MQ, Gao Z, Johnson ATC, Johnson ER, Carpick RW, Martini A (2019) Origin of nanoscale friction contrast between supported graphene, MoS_2 , and a graphene/ MoS_2 heterostructure. *Nano Lett* 19:5496–5505
- Velasco-Soto MA, Pérez-García SA, Alvarez-Quintana J, Cao Y, Nyborg L, Licea-Jiménez L (2015) Selective band gap manipulation of graphene oxide by its reduction with mild reagents. *Carbon* 93:967–973
- Wallace PR (1947) The band theory of graphite. *Phys Rev* 71:622
- Wang X, Long R (2021) Rapid charge separation boosts solar hydrogen generation at the graphene- MoS_2 junction: time-domain Ab initio analysis. *J Phys Chem Lett* 12:2763–2769
- Wang M, Cai L, Wang Y, Zhou F, Xu K, Tao X, Chai Y (2017) Graphene-draped semiconductors for enhanced photocorrosion resistance and photocatalytic properties. *J Am Chem Soc* 139:4144–4151
- Wang S, Li X, Liu Y, Zhang C, Tan X, Zeng G, Song B, Jiang L (2018a) Nitrogen-containing amino compounds functionalized graphene oxide: Synthesis, characterization, and application for the removal of pollutants from wastewater: a review. *J Haz Mater* 342:177–191
- Wang W, Wu Z, Eftekhari E, Huo Z, Li X, Tade MO, Yan C (2018b) High performance heterojunction photocatalytic membranes formed by embedding Cu_2O and TiO_2 nanowires in reduced graphene oxide. *Catal Sci Technol* 8:1704–1711
- Wang M, Sun W, Li M, Wu X, Chen C, Cai T, Zeng Q, Hua Y, Wang L, Xie H (2024a) π -electron injection activated dormant ligands in graphitic carbon nitride for efficient and stable uranium extraction. *J Haz Mater* 478:135445
- Wang X, Zhang J, Wang H, Liang M, Wang Q, Chen F (2024b) Revealing the role of defect in 3D graphene-based photocatalytic composite for efficient elimination of antibiotic and heavy metal combined pollution. *Energy Environ Mater* 7:12616
- Wen Y, Ding H, Shan Y (2011) Preparation and visible light photocatalytic activity of Ag/TiO_2 /graphene nanocomposite. *Nanoscale* 3:4411–4417
- Wen C, Liao F, Liu S, Zhao Y, Kang Z, Zhang X, Shao M (2013) Bifunctional ZnO -RGO-Au substrate: photocatalysts for degrading pollutants and SERS substrates for real-time monitoring. *Chem Commun* 49:3049–3051
- Wu X, Mu F, Zhao H (2020) Recent progress in the synthesis of graphene/CNT composites and the energy-related applications. *J Mater Sci Technol* 55:16–34
- Xiang Q, Yu J (2013) Graphene-based photocatalysts for hydrogen generation. *J Phys Chem Lett* 4:753–759
- Xiang Q, Yu J, Jaroniec M (2012) Graphene-based semiconductor photocatalysts. *Chem Soc Rev* 41:782–796
- Xiao FX, Miao J, Liu B (2014) Layer-by-layer self-assembly of CdS quantum dots/graphene nanosheets hybrid films for photoelectrochemical and photocatalytic applications. *J Am Chem Soc* 136:1559–1569
- Xie Y, Li Z, Liu Y, Ye Y, Zou X, Lin S (2020) Plasmon enhanced bifunctional electro-photo catalytic properties of Pt-Au/graphene composites for methanol oxidation and oxygen reduction reaction. *Appl Surf Sci* 508:145161
- Xiong Z, Zhang LL, Zhao XS (2011) Visible-light-induced dye degradation over copper-modified reduced graphene oxide. *Chem A Euro J* 17:2428–2434
- Xiong T, Ye Y, Luo B, Shen L, Wang D, Fan M, Gong Z (2021) Facile fabrication of 3D TiO_2 -graphene aerogel composite with enhanced adsorption and solar light-driven photocatalytic activity. *Ceram Int* 47:14290–14300
- Xue J, Ma S, Zhou Y, Wang Q (2015) Au-loaded porous graphitic C_3N_4 /graphene layered composite as a ternary plasmonic photocatalyst and its visible-light photocatalytic performance. *RSC Adv* 5:88249–88257
- Yan Y, Manickam S, Lester E, Wu T, Pang CH (2021) Synthesis of graphene oxide and graphene quantum dots from miscanthus via ultrasound-assisted mechano-chemical cracking method. *Ultrasound Sonochem* 73:105519
- Yan C, Liu YL, Zeng Q, Wang GG, Han JC (2023) 2D nanomaterial-supported single-metal atoms for heterogeneous photo/electrocatalysis. *Adv Funct Mater* 33:2210837
- Yang MQ, Xu YJ (2013a) Basic principles for observing the photosensitizer role of graphene in the graphene-semiconductor composite photocatalyst from a case study on graphene-ZnO. *J Phys Chem C* 117:21724–21734
- Yang MQ, Xu YJ (2013b) Selective photoredox using graphene-based composite photocatalysts. *Phys Chem Chem Phys* 15:19102–19118
- Yang MQ, Weng B, Xu YJ (2013) Improving the visible light photoactivity of In_2S_3 -graphene nanocomposite via a simple surface charge modification approach. *Langmuir* 29:10549–10558
- Yang MQ, Zhang N, Pagliaro M, Xu YJ (2014) Artificial photosynthesis over graphene-semiconductor composites: are we getting better? *Chem Soc Rev* 43:8240–8254
- Yang Y, Xu L, Wang H, Wang W, Zhang L (2016) TiO_2 /graphene porous composite and its photocatalytic degradation of methylene blue. *Mater Design* 108:632–639
- Yang X, Rosario-Ortiz FL, Lei Y, Pan Y, Lei X, Westerhoff P (2022) Multiple roles of dissolved organic matter in advanced oxidation processes. *Environ Sci Technol* 56:11111–11131
- Yang Z, Zhou S, Feng X, Wang N, Ola O, Zhu Y (2023) Recent progress in multifunctional graphene-based nanocomposites for photocatalysis and electrocatalysis applications. *Nanomaterials* 13:2028
- Yu W, Li S, Yang H, Luo J (2020) Progress in the functional modification of graphene/graphene oxide: a review. *RSC Adv* 10:15328–15345
- Zhang L, Gao Z, Liu C, Ren L, Tu Z, Liu R, Yang F (2014) N-doped nanoporous graphene decorated three-dimensional CuO nanowire network and its application to photocatalytic degradation of dyes. *RSC Adv* 4:47455–47460
- Zhang N, Yang MQ, Liu S, Sun Y, Xu YJ (2015) Waltzing with the versatile platform of graphene to synthesize composite photocatalysts. *Chem Rev* 115:10307–10377
- Zhang L, Sun L, Liu S, Huang Y, Xu K, Ma F (2016) Effective charge separation and enhanced photocatalytic activity by the heterointerface in MoS_2 /reduced graphene oxide composites. *RSC Adv* 6:60318–60326
- Zhang Y, Guo H, Weng W, Fu ML (2017) The surface plasmon resonance, thermal, support and size effect induced photocatalytic activity enhancement of Au/reduced graphene oxide for selective oxidation of benzylic alcohols. *Phys Chem Chem Phys* 19:31389–31398
- Zhang D, Yang J, Wang J, Yang J, Qiao G (2020a) Construction of Cu_2O -reduced graphene oxide composites with enhanced photoelectric and photocatalytic properties. *Opt Mater* 100:109612

- Zhang J, Wu S, Ma L, Wu P, Liu J (2020b) Graphene oxide as a photocatalytic nuclease mimicking nanozyme for DNA cleavage. *Nano Res* 13:455–460
- Zhang S, Li B, Wang X, Zhao G, Hu B, Lu Z, Wen T, Chen J, Wang X (2020c) Recent developments of two-dimensional graphene-based composites in visible-light photocatalysis for eliminating persistent organic pollutants from wastewater. *Chem Eng J* 390:124642
- Zhang S, Wang H, Liu J, Bao C (2020d) Measuring the specific surface area of monolayer graphene oxide in water. *Mater Lett* 261:127098
- Zhang Q, Xiao Y, Yang L, Wen Y, Xiong Z, Lei L, Wang L, Zeng Q (2023) Branched core-shell a-TiO₂@N-TiO₂ nanospheres with gradient-doped N for highly efficient photocatalytic applications. *Chin Chem Lett* 34:107628
- Zhu M, Chen P, Liu M (2012a) Ag/AgBr/graphene oxide nanocomposite synthesized via oil/water and water/oil microemulsions: a comparison of sunlight energized plasmonic photocatalytic activity. *Langmuir* 28:3385–3390
- Zhu M, Dong Y, Xiao B, Du Y, Yang P, Wang X (2012b) Enhanced photocatalytic hydrogen evolution performance based on Ru-tris(4-carboxyphenyl)pyridine-reduced graphene oxide hybrid. *J Mater Chem* 22:23773–23779
- Zhu M, Li Z, Xiao B, Lu Y, Du Y, Yang P, Wang X (2013) Surfactant assistance in improvement of photocatalytic hydrogen production with the porphyrin noncovalently functionalized graphene nanocomposite. *ACS Appl Mater Interfaces* 5:1732–1740
- Zhu C, Xu J, Song S, Wang J, Li Y, Liu R, Shen Y (2020) TiO₂ quantum dots loaded sulfonated graphene aerogel for effective adsorption-photocatalysis of PFOA. *Sci Total Environ* 698:134275

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Springer Nature or its licensor (e.g. a society or other partner) holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.