

Graphene-Metal Oxide Semiconductor Nanocomposite

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Contributor: Soumen Mandal , Srinivas Mallapur , Madhusudana Reddy , Jitendra Kumar Singh , Dong-Eun Lee , Taejoon Park

Graphene is one of the most favorite materials for materials science research owing to its distinctive chemical and physical properties, such as superior conductivity, extremely larger specific surface area, and good mechanical/chemical stability with the flexible monolayer structure. Graphene is considered as a supreme matrix and electron arbitrator of semiconductor nanoparticles for environmental pollution remediation.

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Aqueous Medium

environmental remediation

1. Introduction

Graphene is a 2-D material composed of layers of carbon atoms crammed into a honeycomb network and has become an escalating star on the prospect of materials science in the past many years [\[1\]\[2\]\[3\]](#). Graphene can be used to produce 0-D fullerene, 1-D and 3-D graphitic carbon nanotubes that had been intensively studied for the last ten years [\[4\]\[5\]](#). Graphene exhibits enthralling assets such as extraordinary conductivity, maximum surface-area-to-volume ratio, a fluorescence-quenching competence by electron or energy-allocation, a quantum Hall effect at room temperature, a bipolar electric field effect laterally with the surface conduction of charge carriers and a tunable band gap [\[6\]\[7\]](#). Narrow band gap metal oxides are of great interest, due to their efficient utilization of solar energy which signifies an auspicious technology to resolve the global energy and eco-friendly challenges [\[5\]\[8\]\[9\]\[10\]](#). Furthermore, graphene sheets decorated with metal oxide nanoparticles exhibit the outstanding properties because of the synergetic effect between them [\[11\]](#).

The growth of graphene-based composites provides a significant milestone to multiply the application enactment of metal oxide nanomaterials in photocatalysis, as the hybrids have adaptable and suitable properties with superior performances over the individual oxide nanomaterials. With keeping this in mind, considerable efforts have been made on decorating graphene with metal oxides [\[12\]](#). Graphene-based materials have also been used as the catalyst in the reactions pertaining to environmental remediation [\[13\]](#). Graphene oxide-based (GO-based)/reduced graphene oxide-based (rGO-based) materials are used as photocatalysts for pollutant abatement [\[14\]\[15\]\[16\]\[17\]](#).

2. Photocatalytic Evaluation

Pristine TiO₂ and ZnO exhibited good photocatalytic activity in UV light due to their wide band gap. These two metal oxides are stable in aqueous conditions during photocatalysis. Further, coupling of graphene with TiO₂ and ZnO increases the photocatalytic activity due to increases in the photogenerated charge carriers. Metal oxides with magnetic properties of metal ferrites (MFe₂O₄) offer an added advantage as photocatalysts since they can be recovered by applying an external magnetic field after catalysis. Metal ferrites (MFe₂O₄, M = Co, Ni, Mn, Zn, etc.) materials are proven to be excellent candidates for visible light photocatalytic H₂ generation through water splitting. Recycling ability for metal ferrites are far better compared to nano semiconductors like TiO₂ and ZnO. MFe₂O₄ is a class of semiconductor with narrow band gap, which exhibits characteristic visible light response, possess good photochemical stability, and exhibits excellent optical properties.

MFe₂O₄ absorbs 42–45% of sunlight, whereas TiO₂ and ZnO absorbs 4% of sunlight. MFe₂O₄ are efficient for the degradation of dye degradation and organic pollutant degradation compared to the other metal oxides (SnO₂, CeO₂, BaTiO₃, and SrTiO₃), with respect to the catalyst and the light source. In MFe₂O₄ context, recombination of photogenerated charge carriers is the major limitation in semiconductor photocatalysis as it reduces the overall quantum efficiency. In order to enhance the photocatalytic activity, graphene material is coupled with MFe₂O₄, where the graphene channels the electrons. Comparison of degradation rate for various photocatalytic reaction systems is incongruous since the nature of catalyst and substrate pollutant molecules are different in each reaction. Ferrite nanoparticles have a strong magnetic property, which can be easily used for magnetic separation after photo-mineralization.

The photocatalytic efficiency depends on the ratio of the photogenerated charge-carrier transfer rate to the rate of electron-hole recombination. For composite structure, M²⁺ ion easily bonds with oxygen by giving an electron and super oxide radical. This super oxide radical can oxidize the organic substrate molecule. The Fe³⁺ ion and Fe²⁺ ions can show photo-Fenton reactions in presence of in-situ-generated H₂O₂. This H₂O₂ generates hydroxyl-free radicals, which are involved in the degradation of pollutants. Predicted mechanism for the rGO-CoFe₂O₄ composite is shown in Equations (1)–(4).

rGO-BiO₆ composite shows better photocatalyst compared to other catalysts prepared from hydrothermal method ([Table 1](#)). The enhanced photocatalytic activity could be endorsed to the negative shift in the Fermi level of graphene-Bi₂WO₆ (G-BWO), decrease the conduction band potential, and elevate migration efficiency of photo-induced electrons, which may restrain the charge recombination efficiently. Superior contact between BiVO₄ and rGO scaffold subsidizes to photo-response augmentation compared to other electrochemical methods in the rGO-BiO₄ composite.

Table 1. Photocatalysts and their typical synthetic methods used for the preparation from GO/rGO-supported composites.

Order	Photocatalyst	Preparation	References
	A. Hydrothermal method for the synthesis of GO/rGO-NCs photocatalyst		

Order	Photocatalyst	Preparation	References
1	rGO-WO ₃	Na ₂ WO ₄ ·2H ₂ O and 0.05 g NaCl were dissolved in the above dispersion and kept stirring for 1 h. The pH was adjusted to 2 by using HCl solution.	[6]
2	rGO-WO ₃	Preset amounts of Na ₂ WO ₄ ·2H ₂ O (100, 200, and 400 mg, respectively) were dissolved in 10 mL above GO suspension. 5 mL 35% HCl was added slowly. Transferred to autoclave heated at 140 °C for 8 h.	[18]
3	rGO_Co ₃ O ₄	GO dispersed into 24 mL of alcohol, sonicating for 60 min in an ultrasonic cleaner. Then, 0.2 M of Co(Ac) ₂ was added to the mixture followed by 1.2 mL of water, and continued to be stirred for 10 h at a temperature of 80 °C. The resulting solution was then transferred into a 40 mL autoclave for hydrothermal reaction at 150 °C for 3 h.	[19]
4	rGO_Co ₃ O ₄	GO dispersed in the Co (C ₂ H ₃ O ₂) ₂ ·4H ₂ O. 10 mL with 28% ammonia solution were added to solution, and transferred into an autoclave for hydrothermal action at 180 °C for 12 h.	[20]
5	rGO/ZnFe ₂ O ₄ -Ag	The composite was synthesized by the co-precipitation of Zn (NO ₃) ₂ ·6H ₂ O, Fe (NO ₃) ₃ ·9H ₂ O, and AgNO ₃ in the presence of the GO powder.	[21]
6	GO-NiFe ₂ O ₄	GO in NiFe ₂ O ₄ was dispersed in deionized water. Then, NiSO ₄ H ₂ O and FeCl ₃ 6H ₂ O (0.02 mol) were dissolved in 15 mL water. Transferred into autoclave and kept under high pressure.	[22]
7	GO-MnFe ₂ O ₄	GO and 60 mL of ethanol with sonication for 1 h and Mn(NO ₃) ₂ solution and Fe (NO ₃) ₃ 9H ₂ O were dissolved. The resulting mixture was transferred into a 100 mL Teflon-lined stainless-steel autoclave and heated to 180 °C for 20 h under autogenous pressure.	[23]
8	rGO-Bi ₂ WO ₆	GO by using Hammer method GO was reduced by ethylene glycol. Bi(NO ₃) ₃ 5H ₂ O was dispersed into 5 mL of 4 M nitric acid solution. Na ₂ WO ₄ 2H ₂ O was dissolved in 5 mL of de-ionized water and then Na ₂ WO ₄ was added dropwise to the solution.	[24]
B. Sol-gel method for the synthesis of GO/rGO-NCs photocatalysts			
9	rGO-TiO ₂	An aqueous solution of Ti(OH) ₄ was added into an aqueous suspension of GO.	[25]
10	rGO-ZnO	An aqueous solution of Zn (AcO) ₂ ·3H ₂ O was added into an aqueous suspension of GO.	[26]
C. Solvothermal technique for the GO/rGO-NCs photocatalysts			
11	GO/CoFe ₂ O ₄ /CdS	Gr-CoFe ₂ O ₄ nanohybrids were sonicated in 60 mL of ethylene glycol for 10 min. The cadmium source containing 0.1431 g of Cd(NO ₃) ₂ 4H ₂ O and 10 mL of ethylene glycol was added to the	[27]

Order	Photocatalyst	Preparation	References
		mixture containing Gr–CoFe ₂ O ₄ nanohybrids. The mixture of 0.0348 g of thiourea, 0.0514 g of PVP, and 10 mL of ethyleneglycol was transferred into the above mixture.	
12	rGO-ZnFe ₂ O ₄	GO dispersed in ZnO _x (OH) _y and FeO _x solutions were put into a 50 mL autoclave.	[28]
D. Colloidal method for the synthesis of GO/rGO-NCs photocatalyst			
13	rGO-ZnFe ₂ O ₄	C ₂ H ₆ O ₂ solution is containing 2M FeCl ₃ 6H ₂ O, and 1M ZnCl ₂ was gradually added. Then, 1MCH ₃ COONa was introduced into the solution and magnetically stirred for 1 h. Then, transferred to autoclave heated at 1800 °C.	[29]
E. Thermal treatment for the synthesis of GO/rGO-NCs photocatalyst			
14	rGO-WO ₃	Na ₂ WO ₄ ·2H ₂ O was dissolved in 30 mL water. Then, nitric acid was added to the solution drop by drop until the precipitate was formed. Dried at 160 °C for 2 h and annealed at 500 °C for 5 h.	[30]
15	rGO-WO ₃	Na ₂ WO ₄ ·2H ₂ O (0.5 g), H ₂ C ₂ O ₄ (1 g), and Na ₂ SO ₄ (4 g) were added into subsequent solution and stirred for 3 h. The pH of the solution was maintained at 1.5 by adding 3M HCl and stirring was continued for 3 h. Then, transferred to autoclave maintained at 180 °C for 24 h.	[31]
16	FGS/ZnO	GO, Zn(NH ₃) ₄ CO ₃ , and poly(vinyl pyrrolidone) as an intermediate to combine zinc with carbon material	[32]
F. Ball-milling method for the synthesis of GO/rGO-NCs photocatalyst			
17	rGO-CoFe ₂ O ₄		[33]
18	rGO-CoFe ₂ O ₄	Co (NO ₃) ₂ ·6H ₂ O and of Fe (NO ₃) ₃ ·9H ₂ O were added to GO (2.5 wt%). The pH is maintained 10	[33]
19	rGO-CoFe ₂ O ₄		[33]
G. Liquid phase deposition method for the synthesis of GO/rGO-NCs photocatalyst			
20	rGO-TiO ₂	TiO ₂ powder (P25, Degussa) was dispersed in deionized water and subsequently added to the graphene oxide solution	[34]
H. Microwave irradiation method for the synthesis of GO/rGO-NCs photocatalyst			
21	rGO-CoFe ₂ O ₄		[35]
22	rGO-CoFe ₂ O ₄	(Co(NO ₃) ₂ ·6H ₂ O and Fe(NO ₃) ₃ ·9H ₂ O and glucose as oxidizer and fuel. GO, nitrates, and glucose were added in water for 30 min ultrasonic treatment.	[35]
23	rGO-CoFe ₂ O ₄		[35]
24	rGO/CoFe ₂ O ₄ /Ag	GO, AgNO ₃ , and CoFe ₂ O ₄ were dissolved in deionized water and stirred for 2 h. Then, solution was further stirred for 2 h under the UV	[36]

Order	Photocatalyst	Preparation	References
		irradiation of a 22 W low-pressure mercury lamp. The product is washed with distilled water and ethanol in an oven at 60 °C for 12 h.	
I. In situ co-precipitation method for the synthesis of GO/rGO-NCs photocatalyst			
25	rGO-ZnO	GO dispersed in aqueous solution containing Zn(CH ₃ COO) ₂ , DMSO, and H ₂ O	[37]
J. Annealing NH₃ atmosphere method for the synthesis of GO/rGO-NCs photocatalyst			
26	rGO/N-TiO ₂	GO and 300 mg of P90 TiO ₂ was added and stirred for 3 h. GO and P90 TiO ₂ and a few drops of tetrabutyltitanate were added.	[38]

Furthermore, the self-redox properties of iron and manganese atoms in MnFe₂O₄ induced by S₂O₈²⁻ were particularly useful for the generation of SO₄⁻. The quenching tests and electron spin resonance (ESR) display that h⁺, O²⁻, SO₄⁻, and OH are accountable for decomposition of antibiotics. Overall, irrespective of other parameters, the solvothermal method is best and helps in crystal growing and super saturation is achieved by reducing the temperature in the crystal growth zone.

Further, noble metal (Ag, Au, Cu, etc.) exhibits surface plasmon resonance (SPR), which is a characteristic feature. The SPR frequency of the metal particles can be tuned into visible light absorption by shifting the size of the deposited metal particles on the catalyst. Deposited metal is involved in multiple crucial roles, such as serving as a passive electron sink with high capacity to store electrons to suppress photogenerated charge carrier recombination, facilitates rapid dioxygen reduction to generate free radicals and direct excitation of metals, especially under visible light, and vectorial electron transfer to the conduction band (CB) of metal oxide. Thereby, showing improvement in the photocatalysis for the removal of various organic pollutants/dyes.

3. Perspectives and Challenges

Graphene nanosheets act as a substrate to support the metal oxides for photocatalytic activity and graphene-based semiconductor photocatalysts are used for environmental remediation. The morphologies of semiconductors, theoretical electronic-structure calculations, and experimental discovery determinations are necessary on GO to persuade the photocatalytic activity, and composition design is an operative method to enhance the photocatalytic properties. Photocatalytic properties depend on the preparative method, and various parameters like initial concentration, oxidant concentration, pH, particle size, number of GO sheets, and source of light should be explored.

The interface regulates the efficacy of the electron-hole separation. Currently, only few methods succeed in unswervingly depicting the interaction of GR and nanoparticles. Atomic force microscopy (AFM), Surface-enhanced Raman scattering (SERS), and scanning transmission electron microscope (STEM) may be the best techniques for determining the interaction of graphene and nanoparticles. Finally, studies on the preparation of a ternary composite as a photocatalyst for both UV and visible-light-driven pollutant photodegradation have been studied

and reported. Especially, for the design of ternary composite, magnetic materials such as Fe, Co, Mn, etc., as a dopant, and possessing unique advantages, show a remarkable photocatalytic activity and photostability.

Further tasks exist in the application of graphene-based composite for the industrial scale. Some innovative applications of the metal oxide-graphene entail specific understanding between the metal oxides and surface of the graphene, which will have a direct impact on the properties of the composite. Designing a structure for the overall photocatalysis process may require further exploiting of GO by chemically modifying methods. A synthetic approach method of GO-based composite structure by using novel materials has not been achieved to date for photocatalysis, but the solutions to the key challenges appear within reach.

In view of this, graphene-based composites possess diverse potential applications, individually having dissimilar desires concerning material properties, and it can be projected that the research on graphene-composite materials will have an optimistic future.

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