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# Water Remediation by G-/GO-Based Photocatalysts

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## **Abstract**

Graphene, a two-dimensional sheet of  $sp^2$  hybridized carbon atoms, has shown to be the most fascinating and promising option among nanomaterials for a variety of applications, because of its unique structure and tunable physiochemical properties. It can be either in the pure form or in its modified derivatives that include graphene oxide (GO), reduced graphene oxide (rGO), graphene-metal nanoparticle composites, graphene-polymer hybrids, and graphene/organic structures that showed improved results while maintaining inherent properties of the material. These modified nanostructures have a variety of applications as catalysts, energy storage/conversion, antimicrobial, and water decontaminant. In the field of environmental science, graphene has been widely used for molecular sieving involving gas phase separation and organic waste removal from water, due to its biocompatibility, various functional groups, and accessible surface area. Modified graphene can also serve as a semiconductor that can increase the efficiency of the photocatalytic ecosystems that results in the inactivation of the microorganisms causing the organic chemicals to degrade.

**Keywords:** graphene, environmental, remediation, photocatalyst, water

## **1. Introduction**

Recently photocatalysis by using semiconductors has fascinated universal consideration for its energy-related and environmental applications. Nevertheless, the decrease in the efficiency of the photocatalysis restricted its practical applications because of the prompt reunion of photogenerated electrons and holes. Thus, to decrease the reunion of charge carriers is significant for improvement of semiconductor photocatalysis. Among numerous approaches, water remediation has been done by rGO-/GO-based materials which are the most favorable candidates due to their high capacity of dye adsorption, prolonged light absorption range, improved separation of charge carriers, and transportation properties leading to improved photoconversion efficiency of the photocatalytic materials [1–74].

### **1.1 Graphene (rGO)-based photocatalysts**

Various numbers of graphene-based photocatalysts have been prepared with its derivatives which mainly comprise metal oxides (e.g., P25 [1, 8],  $TiO_2$  [9–34, 36, 37],  $ZnO$  [17, 39–43],  $CuO$  [44],  $SnO_2$  [13, 45],  $WO_3$  [46]), metals (e.g.,  $Cu$  [51],  $Au$  [52]), metal-metal oxides (e.g.,  $Ag-TiO_2$  [35]), upconversion material—P25 (e.g.,  $YF_3:Yb^{3+}, Tm^{3+}-TiO_2$  [38]), salts (e.g.,  $CdS$  [47–49],  $ZnS$  [50],  $ZnFe_2O_4$  [53],

MnFe<sub>2</sub>O<sub>4</sub> [54], NiFe<sub>2</sub>O<sub>4</sub> [55], CoFe<sub>2</sub>O<sub>4</sub> [56], Bi<sub>2</sub>WO<sub>6</sub> [57–59], Bi<sub>2</sub>MoO<sub>6</sub> [60], InNbO<sub>4</sub> [61], ZnSe [63]), Ag/AgCl [62]), and other carbon material (e.g., CNT [64]).

## 1.2 Graphene oxide (GO)-based photocatalysts

Graphene oxide (GO) has recently received considerable attention due to oxygen-containing functional groups which increase its solubility in solvents for the preparation of GO-based nanocomposites required for photodegradation of pollutants [65–74]. GO-based nanocomposites mainly include metal oxides (TiO<sub>2</sub>) [66–72], metal-free polymers [73], and silver/silver halides [74].

## 2. Preparation of rGO-/GO-based composite photocatalysts

Some of the commonly used synthesis techniques include in situ growth strategy, solution mixing, hydrothermal/ solvothermal, and microwave-assisted process.

### 2.1 In situ growth strategy

This method is usually used to prepare reduced graphene oxide-/graphene oxide-based metal composites. Zhang et al. reported that TiO<sub>2</sub>/graphene composite photocatalyst [14] is synthesized by a simple liquid-phase deposition technique. Moreover, adopting a similar approach, Wang et al. prepared nanocarbon/TiO<sub>2</sub> nanocomposites where titania nanoparticles were decorated by thermal reaction on the surfaces of three different dimensional nanocarbons [9]. While in thermal reduction method, TiO<sub>2</sub>/graphene composite [12] with a remarkable visible light photocatalytic activity was prepared by Zhang et al. using a heat treatment method of GO, where GO changed to reduced graphene oxide. Uniform ZnO nanoparticles were found on functionalized graphene sheets evenly via thermal decay of mixture of zinc salt, graphene oxide, and poly(vinyl pyrrolidone) [39].

Furthermore, Sn<sup>2+</sup> or Ti<sup>3+</sup> ions were converted to oxides at low temperatures, while GO was reduced to reduced graphene oxide by tin or titanium salts in redox method [13–45]. In our recent work, we prepared SnO<sub>2</sub>-G nanocomposite which displayed higher photocatalytic activity in sunlight as compared to bare metal oxide nanoparticles as shown in **Figure 1** [45]. Similarly reduced graphene oxide-zinc oxide composite was prepared where zinc ions were decorated on GO sheets and transformed to metal oxide nanoparticles by using chemical reagents at 150°C. Reduced graphene oxide-ZnO photocatalyst is formed by reducing the graphene oxide [43].

Li et al. prepared uniform mesoporous titania nanospheres on reduced graphene oxide layers via a process of a template-free self-assembly [20]. Du et al. [21] also developed the macro-mesoporous titania-reduced graphene oxide composite film by a confinement of a self-assembly process as shown in **Figure 2**.

Moreover Kim et al. synthesized strongly coupled nanocomposites of layered titanate and graphene by electrostatically derived self-assembly between negatively charged G nanosheets and positively charged TiO<sub>2</sub> nanosols, followed by a phase transition of the anatase TiO<sub>2</sub> component into layered titanate [37]. Chen et al. prepared graphene oxide/titania composites by using the self-assembly technique [72].

While Cu ion-modified reduced graphene oxide [51] prepared by an immersion technique displayed a high photocatalytic activity, gold nanoparticles were decorated on the surface of the reduced graphene oxide through spontaneous chemical reduction of HAuCl<sub>4</sub> by GOR [52] as shown in **Figure 3**.

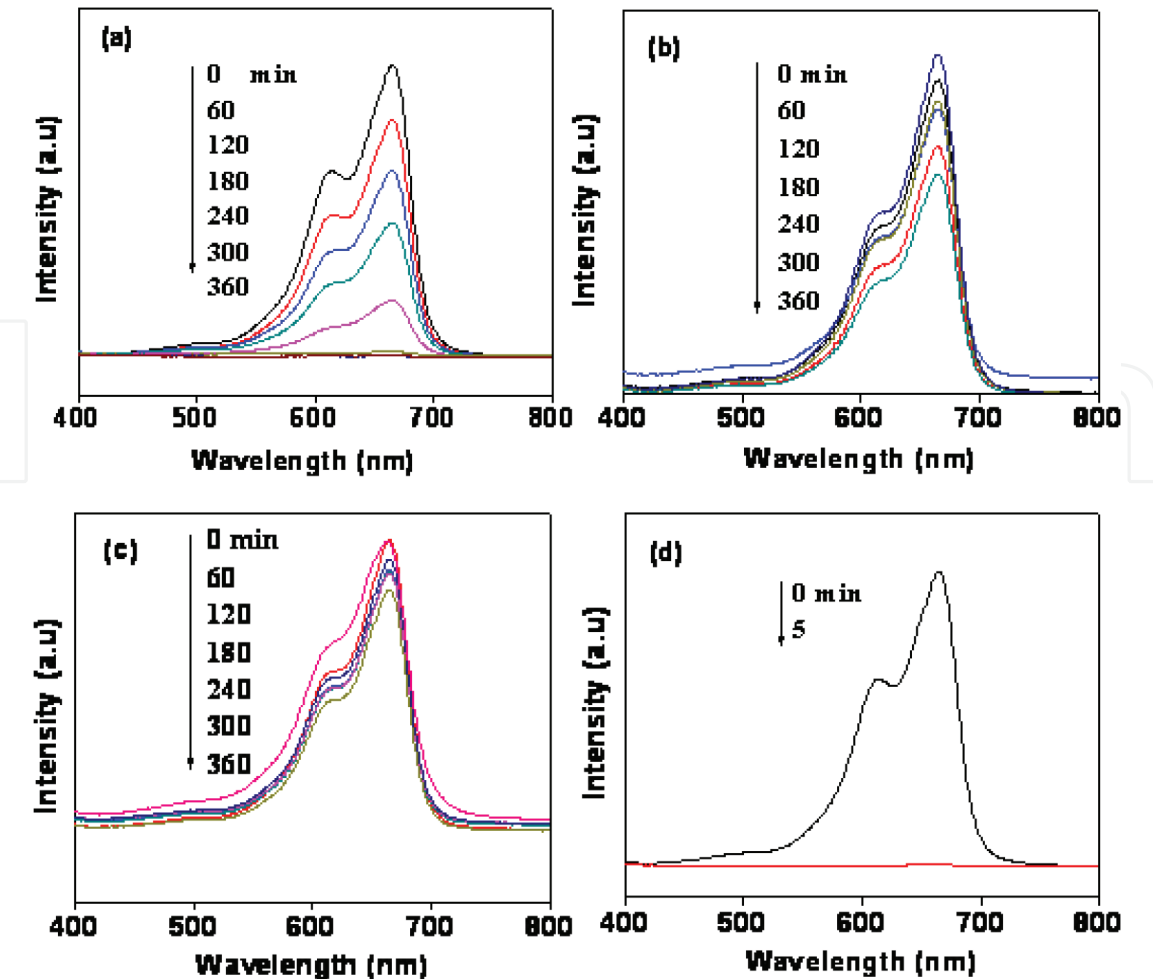


Figure 1.  
Time-dependent absorption spectra of MB solution during UV light irradiation in the presence of (a)  $\text{SnO}_2$  and (b) reduced graphene oxide- $\text{SnO}_2$  and during sunlight irradiation in the presence of (c)  $\text{SnO}_2$  and (d) reduced graphene oxide- $\text{SnO}_2$ . Reprinted with permission of the publisher [45].

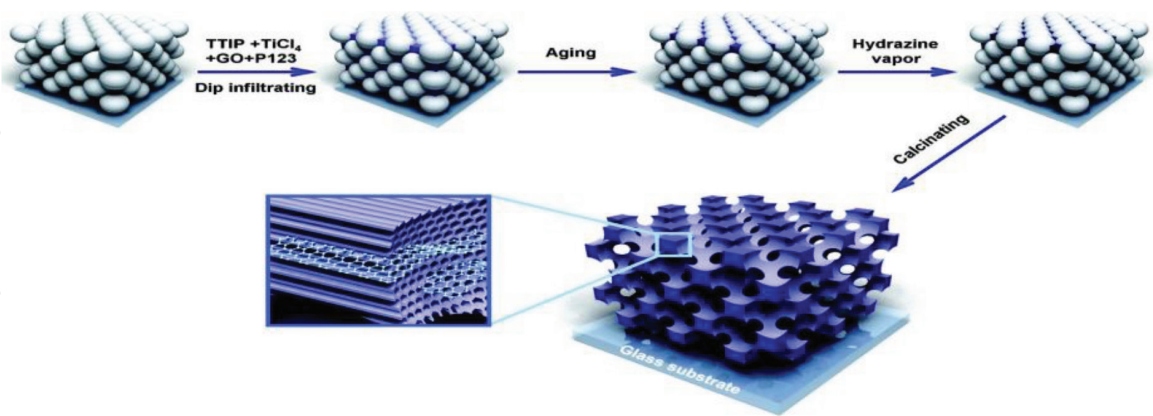
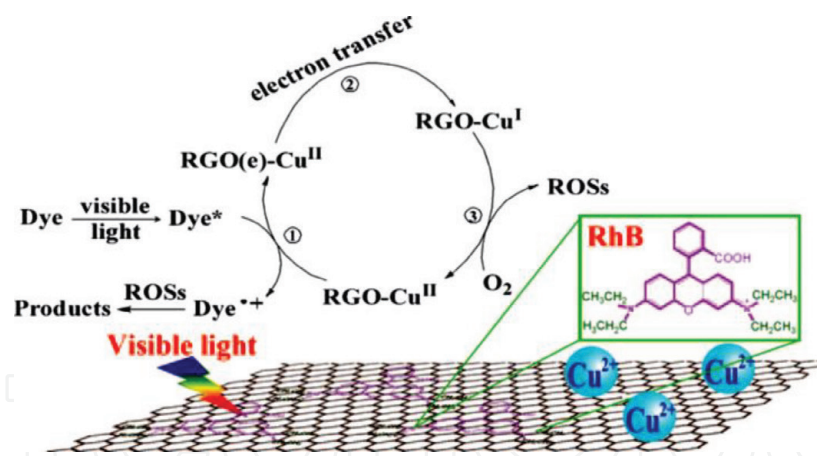


Figure 2.  
Schematic view for the preparation of a macro-mesoporous  $\text{TiO}_2$ -reduced graphene oxide composite film. Reprinted with permission of the publisher [21].

$\text{Bi}_2\text{WO}_6$ /reduced graphene oxide photocatalysts were successfully prepared via in situ refluxing method in the presence of GO [57]. Zhang et al. presented reduced graphene oxide sheet grafted  $\text{Ag@AgCl}$  plasmonic photocatalyst with high activity via a precipitation reaction followed by reduction [62].  $\text{TiO}_2$ -GO was well prepared at  $80^\circ\text{C}$  by using GO and titanium sulfate as precursors [66].

Liu et al. have established a process of water/toluene two-phase for self-assembling  $\text{TiO}_2$  nanorods on graphene oxide [69, 70]. Jiang et al. prepared GO/titania



**Figure 3.**

Possible mechanism of photosensitized degradation of dyes over a rGO Cu composite under visible light irradiation. Reprinted with permission of the publisher [52].

composite by in situ depositing titania on GO through liquid-phase deposition, followed by a calcination at 200°C [71].

GO nanostructures are prepared by modified Hummer's method, which has promising applications in photocatalysis [65].

## 2.2 Solution mixing method

It has been widely used to prepare graphene-based photocatalysts. Previously, titania nanoparticles and GO colloids have been mixed by ultrasonication followed by ultraviolet (UV)-assisted photocatalytic reduction of GO to yield graphene-titania nanocomposites [18, 23, 31].

Akhavan and Ghaderi used a similar strategy to prepare the titania/reduced graphene oxide composite thin film [25].

Guo et al. [28] prepared TiO<sub>2</sub>/graphene composite via sonochemical method. GO/g-C<sub>3</sub>N<sub>4</sub> with efficient photocatalytic capability was also fabricated by the same sonochemical approach [73].

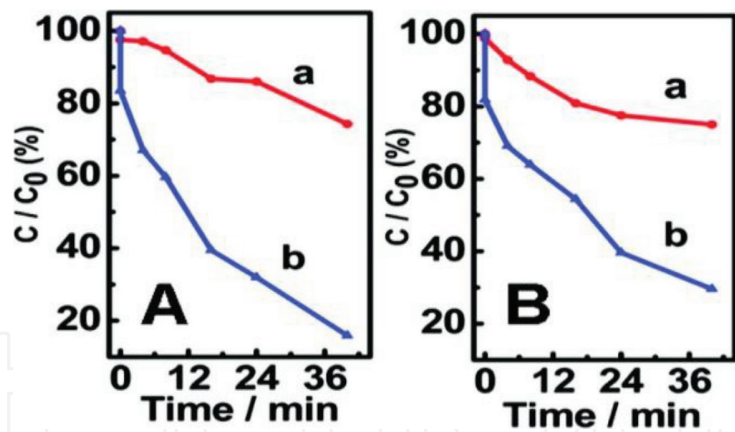
ZnO and GO mixture was dispersed by ultrasonication followed by chemical reduction of GO to graphene ultimately leading to synthesize ZnO/graphene composite [40]. The G-hierarchical ZnO hollow sphere composites are synthesized by Luo et al. by using a simple ultrasonic treatment of the solution [43].

Cheng et al. [40] presented a new facile ultrasonic approach to prepare graphene quantum dots (GQDs), which exhibited photoluminescent in a water solution. The water/oil system is used by Zhu et al. [74] to produce graphene oxide enwrapped Ag/AgX (X = Br, Cl) composites. Graphene oxide and silver nitrate solution were added to chloroform solution of surfactants stirring condition at room temperature to produce hybrid composites which displayed high photocatalytic activity under visible light irradiation as shown in **Figure 4**. Titania/graphene oxide composites were synthesized using one-step colloidal blending method [68].

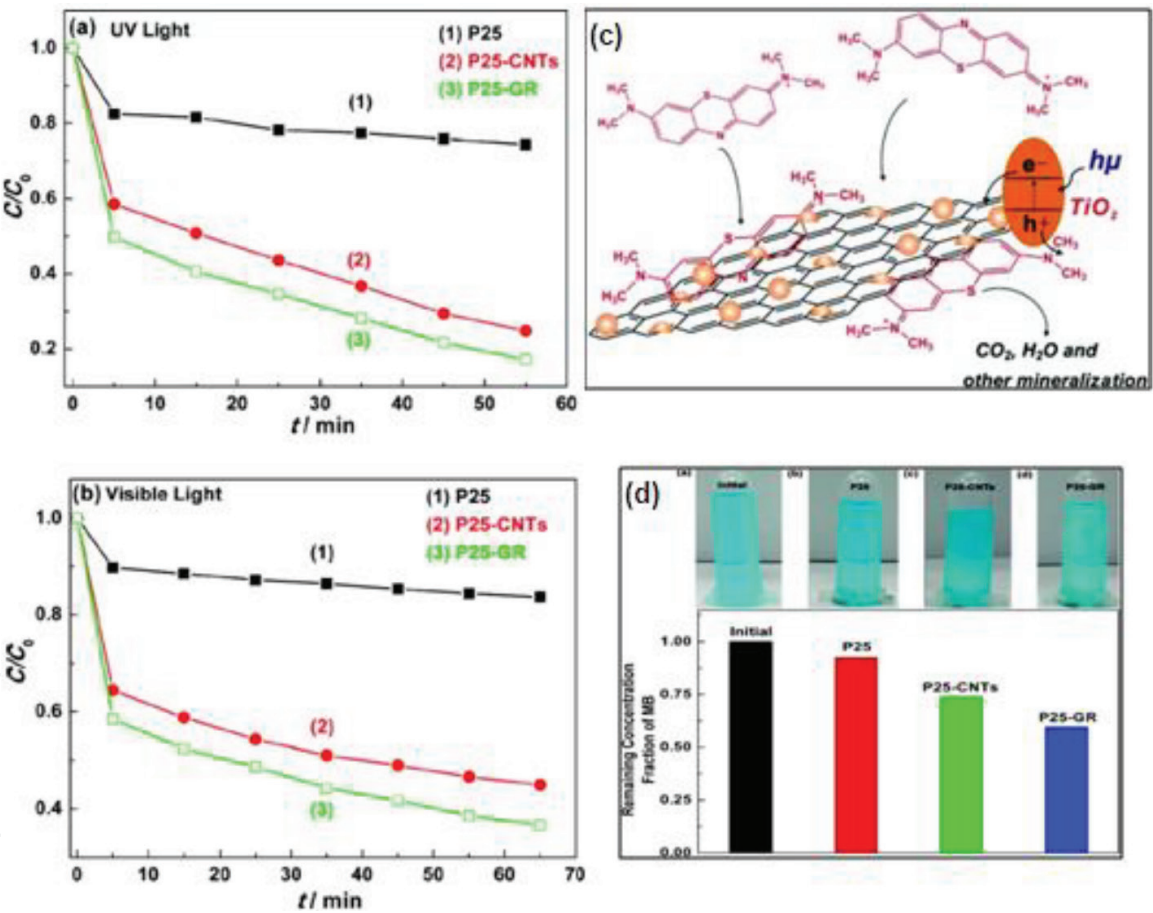
## 2.3 Hydrothermal/solvothermal method

This one-pot process can lead to highly crystalline nanostructures, which operates at elevated temperatures in an autoclave to generate high pressure, without calcination, and at the same time GO reduced to rGO. Typically, graphene-based composites, e.g., P25 [1, 8], TiO<sub>2</sub> [15, 16, 24, 29, 30, 32–34], Ag-TiO<sub>2</sub> [35], UC-P25 [38], WO<sub>3</sub> [46], CdS [49], ZnFe<sub>2</sub>O<sub>4</sub> [53], MnFe<sub>2</sub>O<sub>4</sub> [54], NiFe<sub>2</sub>O<sub>4</sub> [55], Bi<sub>2</sub>WO<sub>6</sub> [58, 59], Bi<sub>2</sub>MoO<sub>6</sub> [60], InNbO<sub>4</sub> [61], and ZnSe [63], have been prepared by the





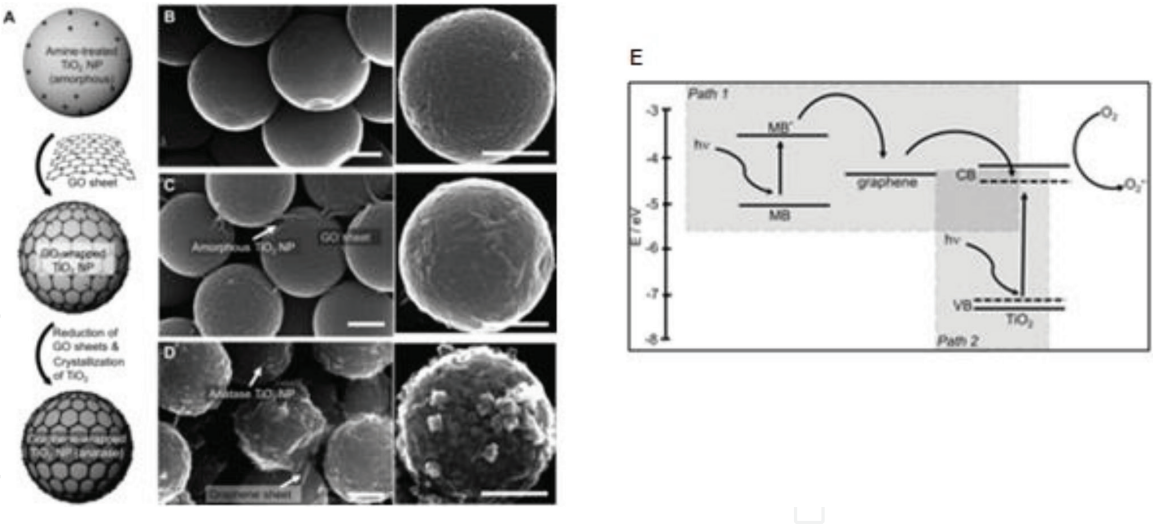
**Figure 4.**  
(A) Photocatalytic activities of silver/silver bromide (a) and silver/silver bromide/GO (b) nanospecies for photodegradation of MO molecules under visible light irradiation and (B) those of the Ag/AgCl (a) and Ag/AgCl/GO (b) nanospecies. Reprinted with permission of the publisher [74].



**Figure 5.**  
Photodegradation of MB under (a) UV light ( $\lambda = 365$  nm) and (b) visible light ( $\lambda > 400$  nm) over (1) P25, (2) P25-CNTs, and (3) P25-GR photocatalysts, respectively. (c) Schematic structure of P25-GR and process of the photodegradation of MB over P25-GR. (d) Bar plot showing the remaining MB in solution: (1) initial and equilibrated with (2) P25, (3) P25-CNTs, and (4) P25-GR in the dark after 10-min stirring. Pictures of the corresponding dye solutions are on the top for each sample. Reprinted with permission of the publisher [8].

hydrothermal process, while others such as TiO<sub>2</sub> [11, 22, 26, 27], CuO [44], CdS [48], and CoFe<sub>2</sub>O<sub>4</sub> [56] are prepared by the solvothermal process.

Li et al. have prepared P25-G nanocomposite using GO and P25 as raw materials via hydrothermal technique [8]. As illustrated in **Figure 5**, the photocatalysis determines that composite showed improved activity toward the photodegradation of methylene blue (MB).

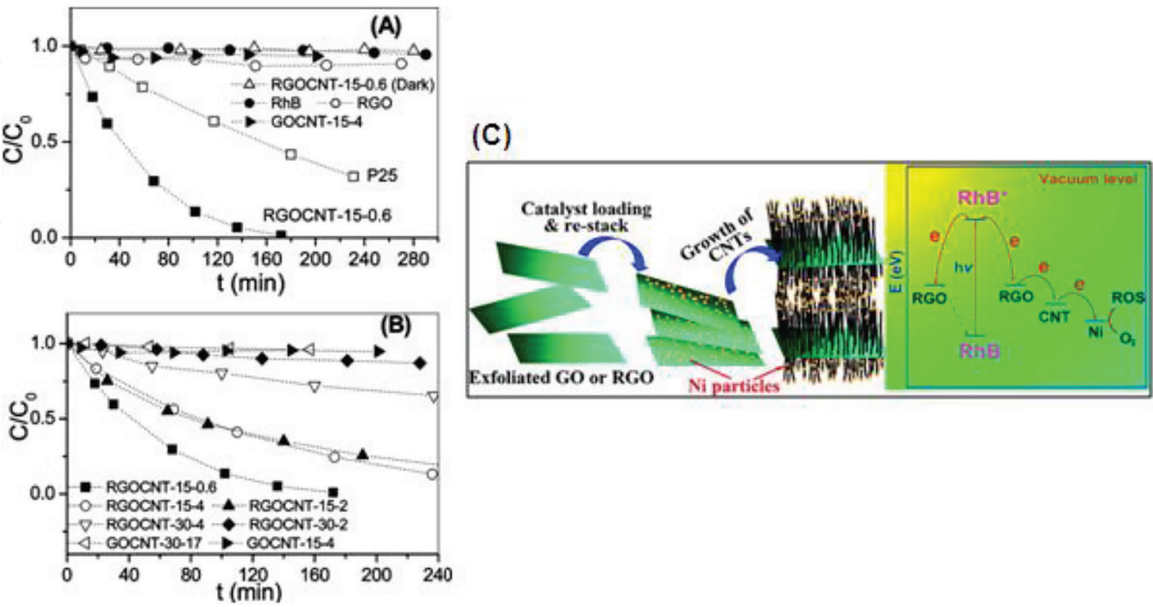


**Figure 6.** (A) Schematic illustration of synthesis steps for graphene-wrapped anatase TiO<sub>2</sub> nanoparticles (NPs) and corresponding SEM images of (B) bare amorphous TiO<sub>2</sub> NPs, (C) GO-wrapped amorphous TiO<sub>2</sub> NPs, and (D) graphene-wrapped anatase TiO<sub>2</sub> NPs (scale bar: 200 nm); (E) the suggested mechanism for the photocatalytic degradation of MB by graphene-wrapped anatase TiO<sub>2</sub> NPs under visible light irradiation. Reprinted with permission of the publisher [8].

Lee et al. synthesized graphene oxide (GO)-wrapped TiO<sub>2</sub> nanoparticles by combining positively charged TiO<sub>2</sub> nanoparticles with negatively charged GO nanosheets, as shown in SEM images in **Figure 6**. Furthermore, it demonstrates the reduction of graphene oxide to reduced graphene oxide and the crystallization of amorphous titania nanoparticles which occurred after a hydrothermal treatment.

2.4 Microwave-assisted method

In situ microwave irradiation is a facile method which has been used for the simultaneous formation of metal oxide (e.g., TiO<sub>2</sub> [17], ZnO [17, 41], CdS [47],



**Figure 7.** (A) Photocatalytic degradation for RhB under different experimental conditions with catalysts GOCNT-15-4 and P25. (B) Photocatalytic properties of different samples in degrading RhB. (C) Experimental steps of pillaring GO and RGO platelets with CNTs while energy diagram showing the proposed mechanism of photosensitized degradation of RhB under visible light irradiation. Reprinted with permission of the publisher [64].

ZnS [50]) and reduction of GO. The drawback of this process is that it did not show its fine control over the uniform size and surface distribution of nanoparticles on G surfaces.

2.5 Other methods

In addition to the abovementioned examples, graphene-based photocatalysts are synthesized by developing new synthetic strategies, e.g., electrospinning [10] and chemical vapor deposition (CVD) [64].

Zhao et al. pillared reduced graphene oxide platelets with carbon nanotubes using the CVD method with acetonitrile as the carbon source and nickel nanoparticles as the catalysts as shown in Figure 7.

Photocatalytic TiO<sub>2</sub> films were prepared by Yoo et al. using RF magnetron sputtering and GO solutions with different concentrations of GO in ethanol which were coated on TiO<sub>2</sub> films [67]. Graphene film was formed on the surface of TiO<sub>2</sub> nanotube arrays through in situ electrochemical reduction of GO dispersion by cyclic voltammetry [19].

3. Photocatalysis

Due to widespread environmental applications, photocatalysis has fascinated an increasing consideration. The graphene-/graphene oxide-based photocatalyst revealed a significant improvement of photocatalytic degradation of methylene blue (MB) [1, 8, 11, 12, 15, 18, 21, 22, 26, 28, 30–33, 35, 36, 40, 41, 43, 45, 48, 50, 52–56, 60, 61, 67–69], rhodamine B (RhB) [13, 20, 24, 27, 32, 42, 44, 51, 52, 56–59, 62, 64, 73], methyl orange (MO) [9, 10, 14, 37, 38, 49, 63, 66, 71, 72, 74], anthracene-9-carboxylic acid (9-AnCOOH) [19], phenol [22, 54], 2,4-dichlorophenoxyacetic acid (2,4-D) [23], 2,4-dichlorophenol [61, 73], malachite green (MG) [29], 2-propanol [34], rhodamine 6G (Rh 6G) [39], rhodamine B 6G (RhB 6G) [46], orange II [52], 2,4-dichlorophenol (2,4-DCP) [61], acid orange 7(AO 7) [64], and resazurin (RZ) [65], as well as photocatalytic reduction of Cr(VI) [17, 47, 71], along with photocatalytic antibacterial activity for killing *E. coli* bacteria [25] by UV [1, 8–10, 13, 14, 16–24, 26, 28–34, 37, 39–45, 50, 54, 65–71], as well as visible irradiation [1, 8–13, 15, 16, 20, 22, 25, 27, 30–33, 35–38, 42, 45–49, 51–64, 67, 68, 72–74], in water which are briefly summarized in Table 1.

| Photocatalysts | Mass fraction | Preparation strategy | Photocatalytic experiments | Performances as compared to reference photocatalyst   | Type of irradiation | References |
|----------------|---------------|----------------------|----------------------------|---|---------------------|------------|
| (1) rGO-based  |               |                      |                            |   |                     |            |
| P25-rGO        | 0.2% G        | Hydrothermal method  | Photodegradation of MB     | 1.17 times higher than P25; DP of 60%   | UV                  | 1          |
|                | 5%            |                      |                            | 1.50 times  |                     |            |
|                | 30%           |                      |                            | 0.97 times  |                     |            |
|                | 0.2%          |                      |                            | 1.42 times higher than P25; DP of 28%   | Visible             |            |
|                | 5%            |                      |                            | 2.32 times  |                     |            |
|                | 30%           |                      |                            | 0.75 times  |                     |            |
| P25-rGO        | 1.0% G        | Hydrothermal method  | Photodegradation of MB     | 340 or 1.21 times higher than P25 or P25-CNTs; DP of 25% or 70%, respectively (2% = 90 min) | UV                  | 2          |



| Photocatalysts        | Mass fraction | Preparation strategy  | Photocatalytic experiments | Performances as compared to reference photocatalyst   | Type of irradiation | References |
|-----------------------|---------------|---|----------------------------|---|---------------------|------------|
| TiO <sub>2</sub> -rGO | 10 mg G       | In situ growth strategy (thermal treatment)                     | Degradation of MO          | 4.33 or 1.18 times higher than P25 or P25-CNTs; DP of 15% or 55%, respectively (2% = 90 min)  | Visible             | 3          |
|                       |               |   |                            | 2.05 times higher than P25; DP of 40%   | UV                  |            |
| TiO <sub>2</sub> -rGO | 0.75% G       | Electrospinning method  | Degradation of MO          | 5.46 times higher than P25; 15%   | Visible             | 4          |
|                       |               |   |                            | 1.51 times higher than TiO <sub>2</sub> ; DP of 54%   | UV                  |            |
| TiO <sub>2</sub> -rGO | 10 mg G       | Solvothermal method   | Photodegradation of MB     | 2.04 times higher than TiO <sub>2</sub> ; DP of approx. 22%   | Visible             | 5          |
|                       |               |   |                            | 2.32 or 1.50 times higher than pure TiO <sub>2</sub> or P25; DP of 25% or 39%, respectively   | Visible             |            |
| TiO <sub>2</sub> -rGO | 10 mg G       | In situ growth strategy (thermal reduction method)              | Photodegradation of MB     | 3.0 or 1.92 times   | Visible             | 6          |
|                       |               |   |                            | 2.88 or 1.84 times  |                     |            |
| TiO <sub>2</sub> -rGO | No data       | In situ growth strategy (redox method)                          | Photodegradation of RhB    | 7.0 times higher than pure P25; DP of 10%   | Visible             | 7          |
|                       |               |   |                            | 1.16 times higher than P25 reaction rate constant = 0.0049 min <sup>-1</sup>  | Visible             |            |
| SnO <sub>2</sub> -rGO |               |   |                            | 0.53 times higher than P25 reaction rate constant = 0.043 min <sup>-1</sup>   | UV                  |            |
|                       |               |   |                            | 2.24 times higher than P25 reaction rate constant = 0.0049 min <sup>-1</sup>  | Visible             |            |
| TiO <sub>2</sub> -rGO | 20 mg G       | In situ growth strategy (simple liquid-phase deposition method) | Photodegradation of MO     | 0.62 times higher than P25 reaction rate constant = 0.043 min <sup>-1</sup>   | UV                  | 8          |
|                       |               |   |                            | 1.89 times higher than P25 and graphene; DP of 45%  | UV                  |            |
| TiO <sub>2</sub> -rGO | No data       | Hydrothermal method   | Photodegradation of MB     | 13.04 or 10.62 times higher than P25 or anatase TiO <sub>2</sub> ; reaction rate constant = 0.0026 min <sup>-1</sup> or 0.0032 min <sup>-1</sup> , respectively | Visible             | 9          |
|                       |               |   |                            | 1.63 times higher than P25; DP of 52%   | UV                  |            |
| TiO <sub>2</sub> -rGO | 20: 1         | Hydrothermal method   | Photodegradation of RhB    |   |                     | 10         |

| Photocatalysts             | Mass fraction | Preparation strategy                              | Photocatalytic experiments  | Performances as compared to reference photocatalyst  | Type of irradiation | References |
|----------------------------|---------------|---|---|--|---------------------|------------|
|                            |               |   |   | 3.33 times higher than P25; DP of 15%  | Visible             |            |
| TiO <sub>2</sub> -rGO      | 0.8% G        | Microwave-assisted method                         | Photocatalytic reduction of Cr(VI)  | 1.09 or 1.30 times higher than pure TiO <sub>2</sub> or commercial P25 = removal rate of 83% or 70%, respectively    | UV                  | 11         |
| rGO-w-TiO <sub>2</sub>     | 1:10          | Solution mixing method                            | Photodegradation of MB  | 1.25 times higher than P25; DP of 80%  | UV                  | 12         |
| TiO <sub>2</sub> -rGO film | No data       | Cyclic voltammetric reduction method              | Photodegradation of anthracene-9-carboxylic acid (9-AnCOOH)               | 2.13 times higher than bare TiO <sub>2</sub> nanotubes; DP of 46%  | UV                  | 13         |
| TiO <sub>2</sub> -rGO      | 6.5% G        | In situ growth strategy (self-assembly synthesis) | Photodegradation of RhB   | 3.92 times higher than TiO <sub>2</sub> ; DP of 25%  | UV-vis              | 14         |
| TiO <sub>2</sub> -rGO      | 0.6% G        | In situ growth strategy (self-assembly method)    | Photodegradation of MB  | 1.57 times higher than TiO <sub>2</sub> ; reaction rate constant = 0.045 min <sup>-1</sup>                           | UV                  | 15         |
| TiO <sub>2</sub> -rGO      | No data       | Solvothermal method                               | Photodegradation of phenol  | 1.68 times higher than P25; DP of 48%  | UV                  | 16         |
|                            |               |   |   | 3.10 times higher than P25; DP of 20%  | Visible             |            |
|                            |               |   | Photodegradation of MB  | 3.5 times higher than P25; DP of 20%   | Visible             |            |
| TiO <sub>2</sub> -rGO film | No data       | Solution mixing method                            | Photodegradation of 2,4-dichlorophen-oxyacetic acid (2,4-D)               | 4.0 times higher than TiO <sub>2</sub> film; reaction rate constant = 0.002 min <sup>-1</sup>                        | UV                  | 17         |
| TiO <sub>2</sub> -rGO      | 10% GO        | Hydrothermal method                               | Photodegradation of RhB   | 4.0 or 2.94 times higher than pure TiO <sub>2</sub> or P25; reaction rate constant = 0.05 or 0.068 min <sup>-1</sup> | UV                  | 18         |
| TiO <sub>2</sub> -rGO      | No data       | Solution mixing method                            | Photocatalytic antibacterial activity for killing <i>E. coli</i> bacteria | 7.55 times higher than TiO <sub>2</sub> ; reaction rate constant = 0.0086 min <sup>-1</sup>                          | Visible             | 19         |
| TiO <sub>2</sub> -rGO      | 0.3 mg GO     | Solvothermal method                               | Photodegradation of MB  | 2.08 times higher than P25; DP of 40.8%  | UV                  | 20         |
| TiO <sub>2</sub> -rGO      | No data       | Solvothermal method                               | Photodegradation of RhB   | 2.79 times higher than P25; reaction rate constant = 0.0162 min <sup>-1</sup>  | Visible             | 21         |
| TiO <sub>2</sub> -rGO      | 75% G         | Sonochemical method                               | Photodegradation of MB  | 2.57 times higher than P25; reaction rate constant = 0.0054 min <sup>-1</sup>  | UV                  | 22         |
| TiO <sub>2</sub> -rGO      | 10% G         | Hydrothermal method                               | Photodegradation of Malachite green                                       | 3.09 times higher than TiO <sub>2</sub> nanotubes; reaction rate constant = 0.0218 min <sup>-1</sup>                 | UV                  | 23         |

| Photocatalysts   | Mass fraction | Preparation strategy                           | Photocatalytic experiments         | Performances as compared to reference photocatalyst  | Type of irradiation | References |
|--|---------------|--|------------------------------------|--|---------------------|------------|
| TiO <sub>2</sub> -rGO  | No data       | Hydrothermal method                            | Photodegradation of MB             | 1.46 times higher than P25; DP of 65%  | UV                  | 24         |
|  |               |  |                                    | 2.41 times higher than P25; DP of 29%  | Visible             |            |
| rGO @TiO <sub>2</sub>  | 1:3           | Solution mixing method                         | Photodegradation of MB             | 4.0 or 1.73 times higher than P25 or physical mixture of G-P25 (1:3); DP of 13% or 30%, respectively | Visible             | 25         |
|  |               |  | Photodegradation of MB             | 2.93–2.20 times higher than P25 or physical mixture of G-P25 (1:3); DP of 30–40%                     | UV                  |            |
| TiO <sub>2</sub> -B-doped rGO  | 2 mg G        | Hydrothermal method                            | Photodegradation of MB             | 4.30 times higher than TiO <sub>2</sub> ; reaction rate constant = 0.010 min <sup>-1</sup>           | UV-vis              | 26         |
|  |               |  | Photodegradation of RhB            | 1.6 times higher than TiO <sub>2</sub> ; reaction rate constant = 0.005 min <sup>-1</sup>            |                     |            |
| TiO <sub>2</sub> -N-doped rGO  |               |  | Photodegradation of MB             | 2.4 times higher than TiO <sub>2</sub> ; reaction rate constant = 0.010 min <sup>-1</sup>            |                     |            |
|  |               |  | Photodegradation of RhB            | 3.2 times higher than TiO <sub>2</sub> ; reaction rate constant = 0.005 min <sup>-1</sup>            |                     |            |
| TiO <sub>2</sub> -rGO-TiO <sub>2</sub>                                 | 0.01 g G      | Hydrothermal method                            | Photodegradation of MB             | 4 times higher than TiO <sub>2</sub>   | UV-vis              | 27         |
| TiO <sub>2</sub> -rGO/MCM-41   | 0.05% G       | Hydrothermal method and Thermal method         | Photodegradation of 2-propanol     | 1.4 times higher than TiO <sub>2</sub> /MCM-41; conversion rate of 26%                               | UV                  | 28         |
|  | 0.15%         |  |                                    | 1.7 times  |                     |            |
|  | 0.4%          |  |                                    | 1.27 times   |                     |            |
|  | 0.6%          |  |                                    | 0.96 times   |                     |            |
| Ag-TiO <sub>2</sub> -rGO   | No data       | Hydrothermal and solution mixing method        | Photodegradation of MB             | Enhancement  | Visible             | 29         |
| RutileTiO <sub>2</sub> -GQD/anatase TiO <sub>2</sub> -GQD              | 0.05 g G      | Solution mixing method                         | Degradation of MB                  | Enhancement for rutile TiO <sub>2</sub> /GQD than anatase TiO <sub>2</sub> /GQD                      | Visible             | 30         |
| Layered titanate rGO   | No data       | In situ growth strategy (self-assembly method) | Photodegradation of MO             | Enhancement as compared to bulk-layered titanates or nanocrystalline-layered titanate                | UV-vis              | 31         |
| UC-P25-rGO<br>UC = YF <sub>3</sub> :Yb <sup>3+</sup> ,Tm <sup>3+</sup> | 4 mg GO       | Hydrothermal method                            | Photodegradation of MO             | 2.88 or times higher than P25 or P25-G or UC-P25; DP of 27% or 53% or 46%, respectively              | Visible             | 32         |
| ZnO-rGO  | 0.6% G        | Microwave-assisted method                      | Photocatalytic reduction of Cr(VI) | 1.12 or 0.92 times higher than pure ZnO or P25; removal rate of 58 or 70%, respectively              | UV                  | 33         |

| Photocatalysts        | Mass fraction | Preparation strategy                                 | Photocatalytic experiments   | Performances as compared to reference photocatalyst   | Type of irradiation | References |
|-----------------------|---------------|--|--|---|---------------------|------------|
| ZnO-FGS               | 0.8% G        | In situ growth strategy (thermal method)             | Photodegradation of Rh 6G  | 1.46 or 1.21 times  | UV                  | 34         |
|                       | 1.0% G        |  |  | 1.68 or 1.40 times  |                     |            |
|                       | 0.1 g GO      |  |  | Enhancement   |                     |            |
|                       | 0.1% G        |  |  | 2.13 times higher than ZnO; reaction rate constant = 0.022 min <sup>-1</sup>  |                     |            |
|                       | 0.5%          |  |  | 2.54 times  |                     |            |
| ZnO-rGO               | 1.0%          | Solution mixing method (sonochemical)                | Photodegradation of MB   | 3.13 times  | UV                  | 35         |
|                       | 2.0%          |  |  | 4.45 times  |                     |            |
|                       | 3.0%          |  |  | 4.13 times  |                     |            |
|                       | 5.0%          |  |  | 3.27 times  |                     |            |
|                       | 1.1% G        |  |  | 1.29 times higher than ZnO; DP of 68%   |                     |            |
| ZnO-rGO               |               | Microwave-assisted method                            | Photodegradation of MB   |   | UV                  | 36         |
| ZnO@ rGO              |               | In situ growth strategy (chemical deposition method) | Photodegradation of RhB  | 1.05 times higher than ZnO; DP of 95%   | UV                  | 37         |
|                       |               |  |  | 1.02 times higher than ZnO; DP of 98%   | Visible             |            |
| ZnO-rGO               | 3.56% G       | Solution mixing method (ultrasonic method)           | Photodegradation of MB   | 2.25 times higher than ZnO; DP of 40%   | UV                  | 38         |
| CuO-rGO               | No data       | Solvothermal method                                  | Photodegradation of RhB in the presence of H <sub>2</sub> O <sub>2</sub> | 2.50 times higher than ZnO; DP of 40%   | UV                  | 39         |
| SnO <sub>2</sub> -rGO | 5% G          | In situ growth strategy (redox method)               | Photodegradation of MB   | 0.40 or times higher than SnO <sub>2</sub> ; DP of 100%   | UV                  | 40         |
|                       |               |  |  | 24.86 times higher than SnO <sub>2</sub> ; DP of 4%   | Visible             |            |
| WO <sub>3</sub> -rGO  | 3.5% G        | Hydrothermal method                                  | Photodegradation of RhB 6G   | 2.2 or 53 times higher than WO <sub>3</sub> nanorods or WO <sub>3</sub> particles; reaction rate constant = 0.00167or 0.000069 min <sup>-1</sup> , respectively | Visible             | 41         |
| CdS-rGO               | 1.5% G        | Microwave-assisted method                            | Photocatalytic reduction of Cr(VI)                                       | 1.16 times higher than CdS = removal rate of 79%  | Visible             | 42         |
| CdS-rGO               | 5% G          | Solvothermal method                                  | Photodegradation of MB   | 2.5 times higher than CdS; DP of 37.6%  | Visible             | 43         |
| CdS-rGO               | 0.01:1        | Hydrothermal method                                  | Photodegradation of MO   | 7.86 times higher than CdS; reaction rate constant = 0.0075 min <sup>-1</sup>   | Visible             | 44         |



| Photocatalysts                        | Mass fraction | Preparation strategy                         | Photocatalytic experiments  | Performances as compared to reference photocatalyst  | Type of irradiation | References |
|---------------------------------------|---------------|--|---|--|---------------------|------------|
| ZnS-rGO                               | No data       | Microwave-assisted method                    | Photodegradation of MB  | 4 times higher than P25; DP of 25%   | UV                  | 45         |
| Cu-rGO                                | No data       | In situ growth strategy (immersion method)   | Photodegradation of RhB   | 2.94 or 30.61 times higher than P25 or graphene; reaction rate constant = $0.0051 \text{ min}^{-1}$ or $0.00049 \text{ min}^{-1}$ , respectively | Visible             | 46         |
| Au-rGO                                | No data       | In situ growth strategy (chemical reduction) | Photodegradation of RhB   | 1.77 times higher than P25; reaction rate constant = $0.0049 \text{ min}^{-1}$   | Visible             | 47         |
|                                       |               |  | Photodegradation of MB  | 8.36 times   |                     |            |
|                                       |               |  | Photodegradation of orange II   | 0.19 times   |                     |            |
| ZnFe <sub>2</sub> O <sub>4</sub> -rGO | 20% G         | Hydrothermal method                          | Photodegradation of MB in the presence of H <sub>2</sub> O <sub>2</sub> | 4.50 times higher than ZnFe <sub>2</sub> O <sub>4</sub> (DP of 22% = 90 min)   | Visible             | 48         |
| MnFe <sub>2</sub> O <sub>4</sub> -rGO | 30% G         | Hydrothermal method                          | Photodegradation of MB  | 9.62 times higher than MnFe <sub>2</sub> O <sub>4</sub> ; DP of 10%  | Visible             | 49         |
|                                       |               |  | Photodegradation of MB  | 1.33 times higher than MnFe <sub>2</sub> O <sub>4</sub> ; DP of 75%  | UV                  |            |
|                                       |               |  | Photodegradation of phenol  | 1.13 times higher than MnFe <sub>2</sub> O <sub>4</sub> ; DP of 75%  | UV                  |            |
| NiFe <sub>2</sub> O <sub>4</sub> -rGO | 25% G         | Hydrothermal method                          | Photodegradation of MB  | Enhancement as compared to NiFe <sub>2</sub> O <sub>4</sub> ; reaction rate constant almost zero (no photocatalytic activity)                    | Visible             | 50         |
| CoFe <sub>2</sub> O <sub>4</sub> -rGO | No data       | Solvothermal method                          | Photodegradation of RhB and MB  | Enhancement  | Visible             | 51         |
| Bi <sub>2</sub> WO <sub>6</sub> -rGO  | 1% G          | In situ growth strategy (refluxing method)   | Photodegradation of RhB   | 1.30 times higher than Bi <sub>2</sub> WO <sub>6</sub> ; DP of 50%   | Visible             | 52         |
|                                       | 2.5%          |  |   | 1.40 times   |                     |            |
|                                       | 5%            |  |   | 1.80 times   |                     |            |
|                                       | 10%           |  |   | 1.10 times   |                     |            |
|                                       | 15%           |  |   | 0.80 times   |                     |            |
| Bi <sub>2</sub> WO <sub>6</sub> -rGO  | 1% G          | Hydrothermal method                          | Photodegradation of RhB   | Enhancement as compared to Bi <sub>2</sub> WO <sub>6</sub>   | Visible             | 53         |
| Bi <sub>2</sub> WO <sub>6</sub> -rGO  | No data       | Hydrothermal method                          | Photodegradation of RhB   | 2.04 times higher than Bi <sub>2</sub> WO <sub>6</sub> ; DP of 44% in 4 min  | Visible             | 54         |
| Bi <sub>2</sub> MoO <sub>6</sub> -rGO | 0.5% G        | Hydrothermal method                          | Photodegradation of MB  | 2.45 times higher than pure Bi <sub>2</sub> MoO <sub>6</sub> ; reaction rate constant $0.0037 \text{ min}^{-1}$                                  | Visible             | 55         |
|                                       | 1%            |  |   | 3.67 times   |                     |            |

| Photocatalysts          | Mass fraction | Preparation strategy                                      | Photocatalytic experiments                 | Performances as compared to reference photocatalyst   | Type of irradiation | References |
|-------------------------|---------------|---|--|---|---------------------|------------|
| InNbO <sub>4</sub> -rGO | No data       | Hydrothermal method                                       | Photodegradation of MB                     | 1.87 times higher than InNbO <sub>4</sub> ; reaction rate constant = 0.0185 min <sup>-1</sup> | Visible             | 56         |
|                         |               |   | Photodegradation of 2,4-dichlorophenol     | 2.10 times higher than InNbO <sub>4</sub> reaction rate constant = 0.0256 min <sup>-1</sup>   |                     |            |
| Ag@AgCl-rGO             | 0.22% G       | Solution mixing method                                    | Photodegradation of RhB                    | 3.88 times higher than Ag@AgCl reaction rate constant = 0.060 min <sup>-1</sup>               | Visible             | 57         |
|                         | 0.44%         |   |  | 4.55 times  |                     |            |
|                         | 1.56%         |   |  | 5.1 times   |                     |            |
| ZnSe-N-doped rGO        | 18 mg G       | Hydrothermal method                                       | Photodegradation of MO                     | Enhancement as compared to ZnSe; (no photocatalytic activity)                                 | Visible             | 58         |
| CNT-rGO                 | No data       | Chemical vapor deposition (CVD) method                    | Photodegradation of RhB                    | 4.28 times higher than P25; reaction rate constant = 0.0049 min <sup>-1</sup>                 | Visible             | 59         |
| (2) GO-based            |               |   |  |   |                     |            |
| GO                      | 1 mg GO       | Solution mixing method (modified Hummers' method)         | Photocatalytic reduction of resazurin (RZ) | No data   | UV                  | 60         |
| TiO <sub>2</sub> -GO    | No data       | In situ growth strategy                                   | Photodegradation of MO                     | 2.27 times higher than pure P25; DP of 38.4%  | UV                  | 61         |
| TiO <sub>2</sub> -GO    | 0.03 mg GO    | RF magnetron sputtering followed by coating               | Photodegradation of MB                     | 2.5 times higher than TiO <sub>2</sub> ; DP of 20%  | UV                  | 62         |
|                         |               |   |  | 1.75 times  | Visible             |            |
| TiO <sub>2</sub> -GO    | 1.2% GO       | Solution mixing method (simple colloidal blending method) | Photodegradation of MB                     | 4.51 times higher than P25 reaction rate constant = 0.0084 min <sup>-1</sup>                  | UV                  | 63         |
|                         | 4.3%          |   |  | 4.98 times  |                     |            |
|                         | 8.2%          |   |  | 8.59 times  |                     |            |
|                         | 1.2%          |   |  | 1.36 times higher than P25 reaction rate constant = 0.0033 min <sup>-1</sup>                  | Visible             |            |
|                         | 4.3%          |   |  | 3.03 times  |                     |            |
|                         | 8.2%          |   |  | 7.15 times  |                     |            |
| TiO <sub>2</sub> -GO    | 50 mg GO      | In situ growth strategy                                   | Photodegradation of MB                     | 1.41 times higher than P25; DP of 70%   | UV                  | 64         |
| TiO <sub>2</sub> -GO    | 500 mg GO     | In situ growth strategy (two phase assembling method)     | Photodegradation of acid orange 7 (AO 7)   | 11.59 times higher than P25 reaction rate constant = 0.0182 min <sup>-1</sup>                 | UV                  | 65         |

| Photocatalysts                      | Mass fraction   | Preparation strategy  | Photocatalytic experiments                      | Performances as compared to reference photocatalyst                           | Type of irradiation | References |
|-------------------------------------|-----------------|---|---|---|---------------------|------------|
| TiO <sub>2</sub> -GO                | No data         | In situ growth strategy (thermal treatment method)  | Photodegradation of MO                          | 7.44 times higher than P25; reaction rate constant = 0.0426 min <sup>-1</sup> | UV                  | 66         |
| TiO <sub>2</sub> -GO                | 0.13% C element | In situ growth strategy (self-assembly method)  | Photodegradation of MO                          | Photocatalytic reduction of Cr(VI)  | Visible             | 67         |
|                                     |                 |   |   | 5.44 times higher than P25; conversion rate = 0.0127 min <sup>-1</sup>        |                     |            |
|                                     |                 |   |   | 1.18 times higher than pure P25; DP of 22%                                    |                     |            |
|                                     |                 |   |   | 0.14%   |                     |            |
|                                     |                 |   |   | 1.59 times  |                     |            |
|                                     |                 |   |   | 0.25%   |                     |            |
|                                     |                 |   |   | 1.0 times   |                     |            |
|                                     |                 |   |   | 0.51%   |                     |            |
|                                     |                 |   |   | 0.82 times  |                     |            |
| g-C <sub>3</sub> N <sub>4</sub> -GO | 1 g GO          | Solution mixing method (sonochemical method)  | Photodegradation of RhB and 2,4-dichloro-phenol | 1.90 times higher than g-C <sub>3</sub> N <sub>4</sub> ; DP of 49.5%          | Visible             | 68         |
| Ag/AgCl/GO                          | No data         | Solution mixing method (surfactant-assisted assembly protocol via an oil/water microemulsion) | Photodegradation of MO                          | 2.84 times higher than Ag/AgCl; DP of 25%                                     | Visible             | 69         |
| Ag/AgBr/GO                          |                 |   | Photodegradation of MO                          | 3.40 times higher than Ag/AgBr; DP of 25%                                     | Visible             |            |

**Table 1.**  
*Photocatalytic degradation of pollutants.*

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