



Review

A Comprehensive Review of Graphitic Carbon Nitride (g-C₃N₄)–Metal Oxide-Based Nanocomposites: Potential for Photocatalysis and Sensing

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Abstract: g-C₃N₄ has drawn lots of attention due to its photocatalytic activity, low-cost and facile synthesis, and interesting layered structure. However, to improve some of the properties of g-C₃N₄, such as photochemical stability, electrical band structure, and to decrease charge recombination rate, and towards effective light-harvesting, g-C₃N₄–metal oxide-based heterojunctions have been introduced. In this review, we initially discussed the preparation, modification, and physical properties of the g-C₃N₄ and then, we discussed the combination of g-C₃N₄ with various metal oxides such as TiO₂, ZnO, FeO, Fe₂O₃, Fe₃O₄, WO₃, SnO, SnO₂, etc. We summarized some of their characteristic properties of these heterojunctions, their optical features, photocatalytic performance, and electrical band edge positions. This review covers recent advances, including applications in water splitting, CO₂ reduction, and photodegradation of organic pollutants, sensors, bacterial disinfection, and supercapacitors. We show that metal oxides can improve the efficiency of the bare g-C₃N₄ to make the composites suitable for a wide range of applications. Finally, this review provides some perspectives, limitations, and challenges in investigation of g-C₃N₄–metal-oxide-based heterojunctions.



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Keywords: graphitic carbon nitride; g-C₃N₄; metal oxide; photocatalysts; sensors; bacterial disinfection; supercapacitors

1. Introduction

Graphitic carbon nitride (g-C₃N₄) is a polymeric, visible-light-active photocatalyst with a bandgap of ~2.7 eV (~460 nm), that was introduced since 2006 [1]. g-C₃N₄ has become an important material in chemistry, physics and engineering because of its facile, low-cost, environmentally-friendly preparation methods with promising stability and good physicochemical properties for use in a wide range of applications [2]. Compared with other semiconductors, g-C₃N₄ can be easily synthesized by various methods with desirable electrical structures as well as morphologies, and high thermal stability up to 600 °C in the air [3,4].

The most common precursors used to prepare g-C₃N₄ are melamine, dicyandiamide, cyanamide, urea, thiourea, and ammonium thiocyanate. Among different types of carbon nitrides such as α-C₃N₄, β-C₃N₄, cubic C₃N₄, pseudocubic C₃N₄, with bandgaps of around 5.49 eV, 4.85 eV, 4.30 eV, and 4.13 eV, respectively, g-C₃N₄ is the most stable phase under ambient conditions [2]. In order to enhance the performance and modulate the properties of g-C₃N₄, researchers have proposed different methods such as doping and making heterojunction with other materials. Examples of these materials are metal oxides, metal sulfides, noble metals, and carbonaceous nanomaterials [5–12]. Among them, metal oxides are the most common ones to improve the efficiency of g-C₃N₄, e.g., increasing the light absorption and reducing the recombination of electrons and holes by promoting the separation of charge carriers. This is mainly due to their suitable band structures [13–18].

The g-C₃N₄ structure has been widely used in many applications, in particular in energy-related applications. Energy consumption to provide electricity and heat will rise

to twice its current consumption by 2050, which is mainly due to industrialization, urbanization, and population growth [19,20]. The consumption of fossil fuels, such as natural gas, coal, and oil, should be decreased as their usage results in detrimental environmental impacts [2,19,20]. Two remedies are solar energy and photocatalysis [21–24]. Both require suitable semiconductors such as $g\text{-C}_3\text{N}_4$ with superior activities for different catalytic reactions, such as organic pollutants degradation, H_2 , and O_2 generation by water splitting and CO_2 reduction to hydrocarbon fuels [14,25–29]. The $g\text{-C}_3\text{N}_4$ can also be used for water disinfection and bacterial control [25,30].

It is evident from Figure 1 that the number of publications, collected from the Scopus database, has been growing fast from 2012 in the field of $g\text{-C}_3\text{N}_4$ and $g\text{-C}_3\text{N}_4$ –metal oxide-based heterojunctions. Figure 1a shows the number of all publications on $g\text{-C}_3\text{N}_4$ since 2012, showing that this topic is among the hot research areas. It is therefore important to provide a comprehensive review of the $g\text{-C}_3\text{N}_4$ –metal oxide composites. To the best of our knowledge, no publication reviews a wide range of papers to explain the applications and structure of these heterojunctions comprehensively.

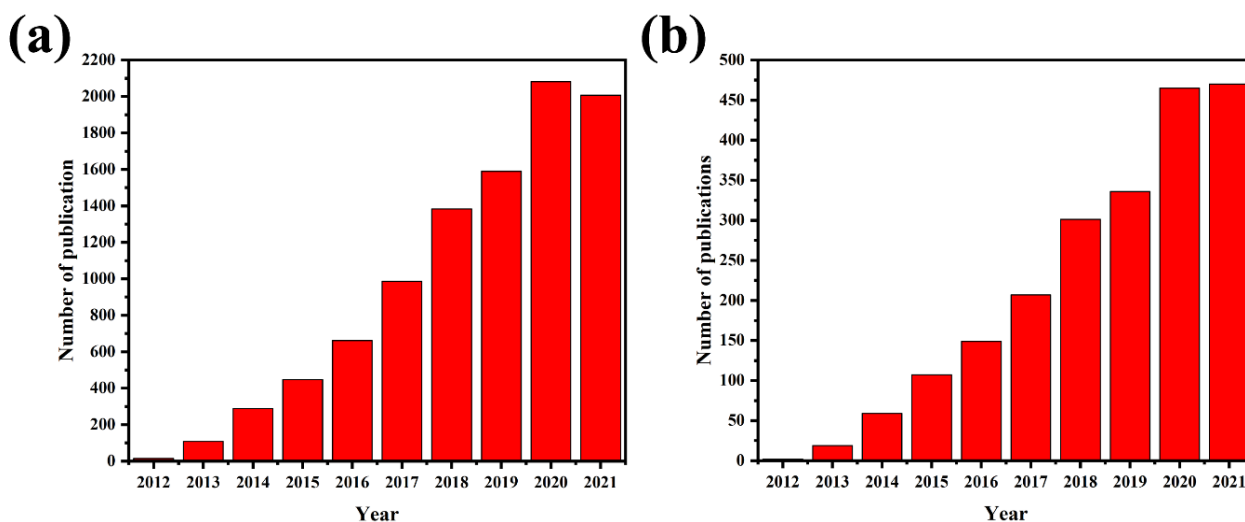


Figure 1. Number of annual publications (a) using “ $g\text{-C}_3\text{N}_4^*$ ” as a keyword since 2012, (b) using “ $g\text{-C}_3\text{N}_4^*$ ” with metal oxides (“ TiO_2 ”, “ ZnO ”, “ WO_3 ”, “Iron Oxide”, “Tin Oxide”, and other metal oxides) as keywords since 2012. Adapted from Scopus database, dated 1 October 2021.

This review covers the research up to 1 October 2021. We highlighted some general information about the structure and characterization of the bare $g\text{-C}_3\text{N}_4$. We also discussed some modifications such as doping to improve the $g\text{-C}_3\text{N}_4$ properties. As well, we have also summarized the research on modification of the structure and properties, to enhance the efficiency of $g\text{-C}_3\text{N}_4$ for different applications, via combining $g\text{-C}_3\text{N}_4$ with metal oxides such as TiO_2 , ZnO , iron oxide, WO_3 , and tin oxide. After reviewing $g\text{-C}_3\text{N}_4$ –metal oxides, we focus on the applications of these kinds of heterojunctions. In the last part of this review article, we suggest some potential investigations for the future in this field that have not been conducted to this date to our knowledge. We also recommend some studies to better understand the nature of these heterojunctions. Most graphs in this review are reproduced by us from the data in original research papers cited, unless stated otherwise in the captions where the permission has been obtained.

2. Structure and Properties of $g\text{-C}_3\text{N}_4$

Melamine, melam, melem, and melon are recognized as heptazine- and triazine-based molecular compounds (coplanar tri-*s*-triazine unit as the elementary structural motif of $g\text{-C}_3\text{N}_4$ structure) to prepare $g\text{-C}_3\text{N}_4$. As illustrated in Figure 2, triazine (C_3N_3) and tri-*s*-triazine/heptazine (C_6N_7) rings are the basic tectonic units of $g\text{-C}_3\text{N}_4$.

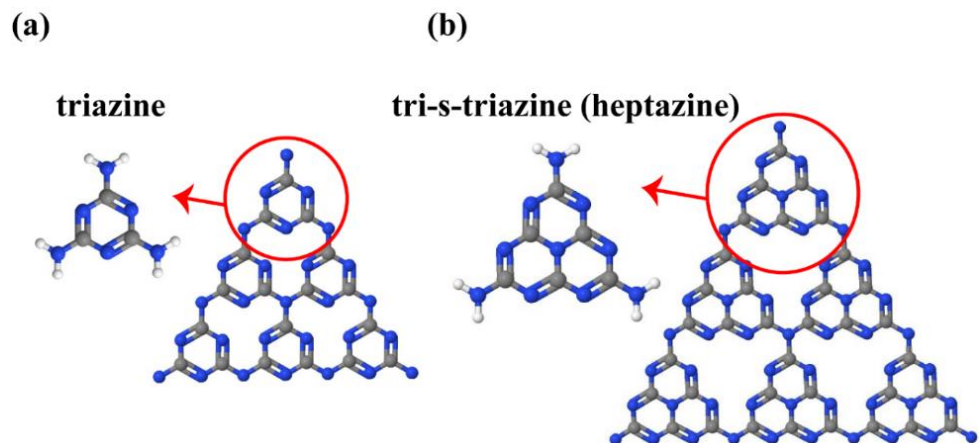


Figure 2. (a) Triazine and (b) tri-s-triazine (heptazine) structures of $g\text{-C}_3\text{N}_4$ (gray, blue, and white balls are carbon, nitrogen, and hydrogen, respectively).

Yan et al. properly studied the phase transition during heating of melamine from room temperature to $1000\text{ }^\circ\text{C}$ with a heating rate of $10\text{ }^\circ\text{C}/\text{min}$ [31]. Figure 3a shows that melamine sublimation and thermal condensation occur at 297 to $390\text{ }^\circ\text{C}$, observed from significant endothermic differential scanning calorimetry (DSC) peak and drastic thermogravimetric analysis (TGA) weight loss. Other endothermic peaks at 545 and $630\text{ }^\circ\text{C}$ are attributed to the materials' deamination and decomposition, respectively.

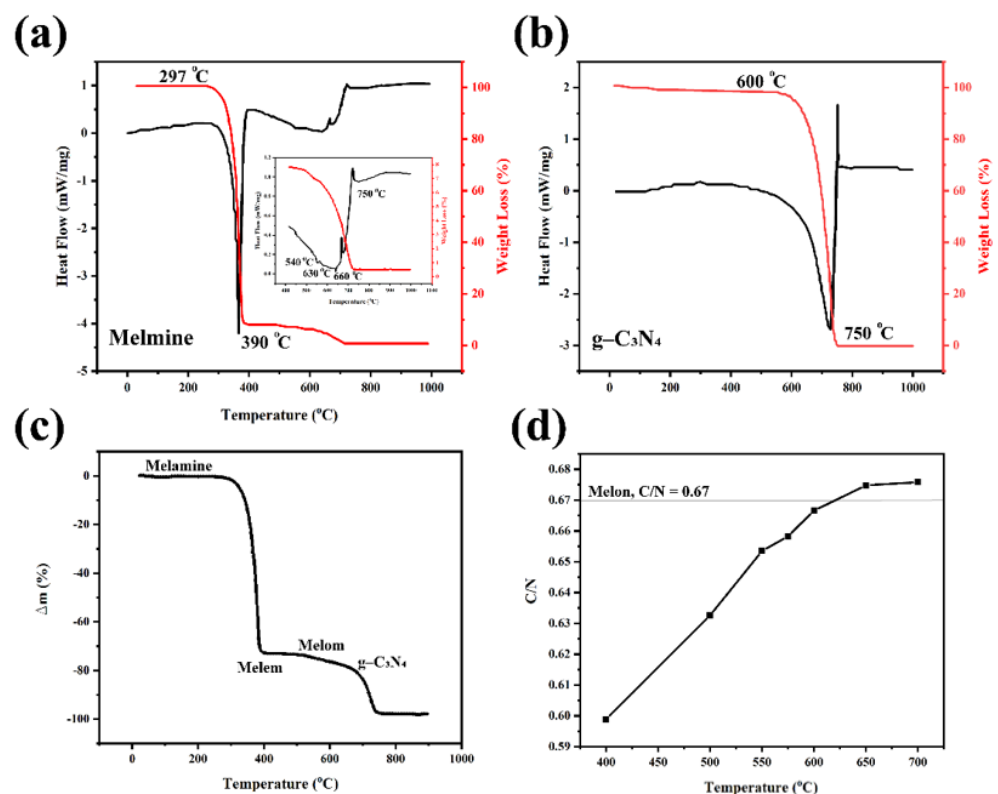


Figure 3. (a) Melamine and (b) $g\text{-C}_3\text{N}_4$ thermal analyses, Copyright © 2022 American Chemical society [31]; (c) thermogravimetric curve of annealing of melamine; (d) C/N values of melamine products at different temperatures, Copyright © 2022 Elsevier [32].

Figure 3b shows the $g\text{-C}_3\text{N}_4$ thermal stability and phase transitions in an open system. This figure shows that $g\text{-C}_3\text{N}_4$ has high stability below $600\text{ }^\circ\text{C}$, which is $30\text{ }^\circ\text{C}$ lower than

the melamine decomposition temperature (630 °C). Beyond this temperature, g-C₃N₄ starts to decompose into small molecules (e.g., CO₂ and NH₃) [31]. The stability of g-C₃N₄ is higher in the semiclosed ammonia atmosphere than in the open system, mainly due to the inhibition of deamination under ammonia atmosphere. The TGA result illustrated that there is no residue at 750 °C [31]. The annealing temperature is a vital factor in the preparation and properties of the final g-C₃N₄ structure. Praus et al. used melamine as a precursor to synthesize g-C₃N₄ in an air atmosphere [32]. The TGA results, illustrated in Figure 3c, demonstrates that by heating the melamine to 400 °C melon is formed. The main reason for the weight loss at this temperature range is the elimination of ammonium. By further heating up to 600 °C, melon is obtained by melon polymerization [33]. In other words, at higher temperatures, the g-C₆N₉H₂ or g-C₃N_{4.5}H is more stable than g-C₃N₄. Figure 3d illustrates that the C/N molar ratio is temperature dependent. The main difference in the C/N value of the synthesized g-C₃N₄ compared to the theoretical value of 0.75 for g-C₃N₄ is due to the incomplete condensation of the amino groups of melon and the low degree of polymerization. Reaction atmosphere is another factor affecting the g-C₃N₄ by providing defects and disordered structures. For instance, in the H₂ atmosphere, by dicyanamide thermal condensation, more nitrogen vacancies were formed [34].

3. g-C₃N₄ Characterizations

The presence of X-ray diffraction (XRD) peaks at about 13 and 27° is an indication of the formation of g-C₃N₄, corresponding to the (002) and (100) diffraction patterns, respectively [35–37]. Paul et al. showed the effects of calcination temperature on the naked g-C₃N₄ using XRD data, Fourier transform infrared spectra (FT-IR), bandgap structure, Brunauer–Emmett–Teller (BET), and photocatalytic recyclability data [36]. Due to the polycondensation of melamine at lower than 400 °C, the crystallinity of the g-C₃N₄ structure is detected (Figure 4a). In contrast, Figure 4b demonstrates that no significant differences are revealed in FTIR spectra at different calcination temperatures. In the FTIR spectra, the characteristic bands with high intensity at the wavenumber about 1640, 1569, 1412, 1328, and 1240 cm⁻¹ are related to the stretching modes of C=N and C–N heterocycles. Besides, the strong peaks at 815 cm⁻¹ are attributed to the s-triazine units. The broad range peak between 3000 cm⁻¹ and 3500 cm⁻¹, is due to the N–H stretching and the remaining water molecules in the structure [35–39]. The BET specific surface areas were estimated to be 37.8, 73.7 and 65.6 m²·g⁻¹ for g-C₃N₄ synthesized at the calcination temperature of 450, 550 and 650 °C, respectively. Calcination temperature also affects the g-C₃N₄ bandgap. The bandgap when calcinated at 550 °C is narrower than when calcinated at 450 and 650 °C (Figure 4c) [36,38,40]. When the temperature increases to 700 °C, a higher bandgap is observed. In other words, the higher the degree of g-C₃N₄ polymerization, the larger the π-plane conjugation degree of heptazine rings via N₂ atoms, at the higher temperature.

To assess the catalytic activity, ultraviolet-visible (UV-vis) diffuse reflectance spectrums (DRS) of the materials were determined at different temperatures [36]. As discussed earlier the results show that the bandgap of g-C₃N₄ prepared at 550 °C is narrower than those formed at 450 and 650 °C, so the structure prepared at the 550 °C absorbs more visible light, which can lead to the more appealing photoactivity. Moreover, the adsorption and degradation efficiencies for methylene blue dye by g-C₃N₄ prepared at 500 °C is 34.4% and 62.6%, which is higher than those synthesized at 450, 550, 600, and 650 °C [36]. Figure 4d reveals that the prepared g-C₃N₄ at a calcination temperature of 550 °C also showed good stability even after four cyclic runs. The pH, and catalyst loading are also important factors in the adsorption and degradation efficiency [36].

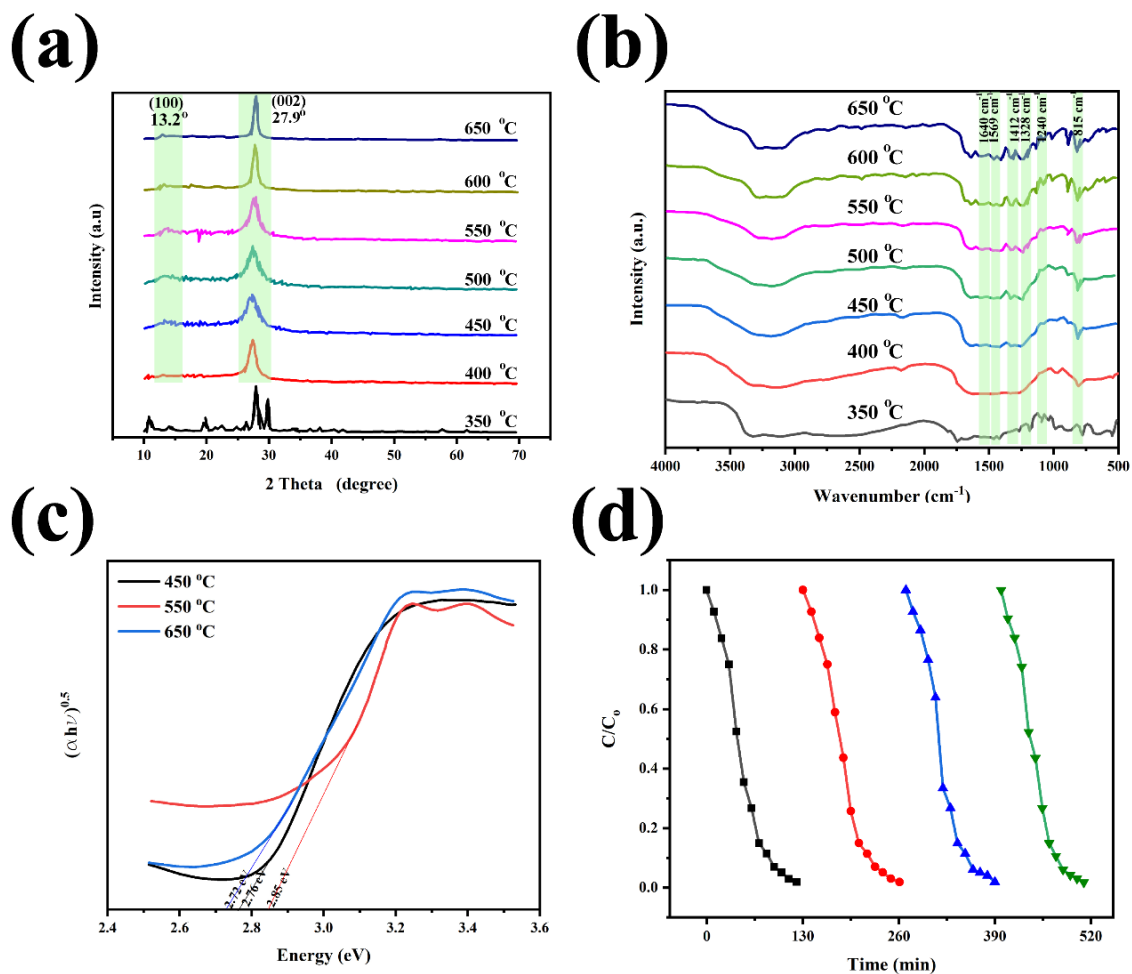


Figure 4. (a) XRD, (b) FTIR, (c) bandgaps of g-C₃N₄ prepared at different calcination temperatures, and (d) the photocatalytic recyclability degradation of dyes using prepared g-C₃N₄ at 550 °C [36].

Chen and coworkers illustrated that the PL depends on the condensation temperature (Figure 5a), showing the strong and stable emission in the range of 450–510 nm, mainly due to the $\pi^*-\pi$, and $\pi^*-\text{LP}$ transitions [41]. The increase in the amount of the tri-s-triazine content at high temperature leads to the higher π states and cause orbital overlap and reduces the PL intensity. The UV-Vis spectra of the g-C₃N₄ at different synthesis temperatures are illustrated in Figure 5b. Like the PL emission peak, the strong absorption peaks at 450 nm and 500 nm are due to the $\pi^*-\pi$ and $\pi^*-\text{LP}$ transitions. Another study, conducted by Dias et al., concluded that during thermal treatment of g-C₃N₄, the enhanced porosity is mainly due to the creation of N vacancies and defects/holes within the nanosheets, resulting in the improvement of optoelectrical properties. Not only the treatment leads to the appearance of $n \rightarrow \pi^*$ transitions, but we can also observe red shift in the absorption spectra. Moreover, it can enhance the photocatalytic activities by improving the carrier separation, reducing charge recombination, which is mainly due to the presence of trap states [42]. Furthermore, researchers also investigated the effect of the different dopants on the absorption properties of g-C₃N₄ [43]. These results showed the effect of temperature and dopants on the optical properties of g-C₃N₄. Yuan et al. showed the photograph of the melamine and g-C₃N₄ synthesized at different temperatures, graphically showing the color variation in the deionized water under UV light (365 nm) (Figure 5c) [44]. By increasing the synthesis temperature, the emitted color varied from blue–violet to green, and the intensity went through a maximum at 550 °C. The impact of the temperature on the PL spectra was demonstrated by the 5–10 nm blue-shift at lower temperatures, due to a less delocalized orbital resulting in a larger bandgap. Figure 5d also reveals strong PL emission spectra

with a much smaller blue-shift. This research is among the rare studies investigating the effect of the environment on PL emission spectra. The g-C₃N₄ time-resolved PL spectra shows 5 ns electron-hole recombination at 25 °C [45].

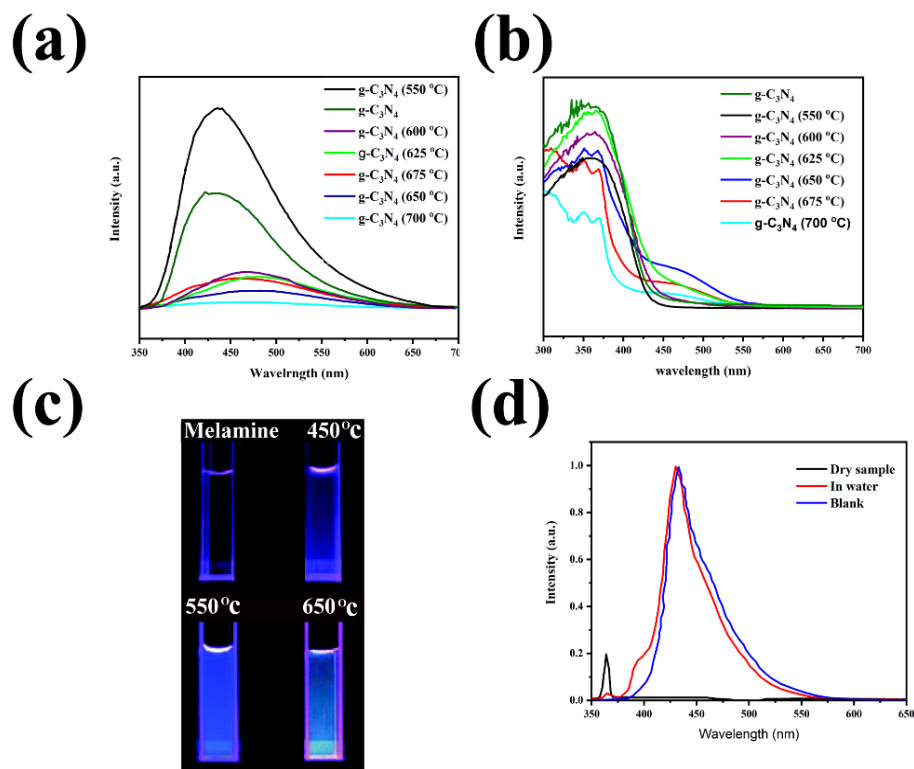


Figure 5. (a) The PL spectra under 325 nm excitation (b) UV-Vis spectra of g-C₃N₄, prepared at different temperatures, Copyright © 2022 American Chemical Society [41]; (c) The g-C₃N₄ in deionized water under 365 nm light; (d) the PL spectra of the g-C₃N₄, g-C₃N₄ in water and in the DI water [44].

4. g-C₃N₄ Preparation

Synthetic techniques for graphitic carbon nitride (g-C₃N₄) were first reviewed by Thomas et al. in 2008 [26]. g-C₃N₄ can be synthesized with various techniques such as chemical vapor deposition (CVD), solvothermal, and plasma sputtering deposition. Chemical vapor deposition to deposit g-C₃N₄ thin film on indium-tin-oxide (ITO), was performed by Ye et al. [27]. In this method, the mixture of thiourea and melamine was put at the bottom of a crucible with ITO substrate above it and finally transferred to the muffle furnace. CVD has several advantages and disadvantages. To be more specific, thin films prepared by CVD are cohesive in all dimensions, which is suitable for elaborately shaped pieces and helps users to fill the insides, undersides, high aspect ratio holes, etc. CVD does not require high vacuum and can deposit a wide variety of materials to prepare high purity composites. In contrast, physical vapor deposition (PVD) such as sputtering requires a high vacuum atmosphere. The drawback of CVD is that some CVD precursors are costly, and can be highly toxic, explosive, or corrosive, such as Ni(CO)₄, B₂H₆, and SiCl₄, respectively. The by-product of this method, including CO, or HF, can also be hazardous. The substrates are limited since they should tolerate high temperatures [46].

The graphitic carbon nitride nanocone arrays were grown onto the Ni-coated Si (100) substrate, using plasma sputtering deposition [47]. This method requires a vacuum chamber and a plasma source with a discharge cavity. Thermal condensation is an economical, energy-efficient approach, which has a higher chance for scaleup for commercialization. The solvothermal methods have some drawbacks such as more synthetic steps compared

to the thermal condensation method; however, they can be cost-effective, low energy consumption methods with more controllable size and morphology [48–50].

Several C–N precursors such as urea, thiourea, melamine, cyanamide, and dicyanamide are used for g-C₃N₄ synthesis (Figure 6a). The procedure is depicted in Figure 6b [51–55]. Among them, cyanamide and its derivatives such as dicyanamide suffer from low solubility and high cost [56]. Pham and Shin showed that urea and melamine cause poor interconnection of g-C₃N₄ to NiTiO₃ since melamine provides a segregated g-C₃N₄ structure with no connection to the NiTiO₃ phase, and urea makes a condensed g-C₃N₄ structure by releasing oxygen-containing gas during the thermal condensation [56]. Dicyandiamide and thiourea with higher reactivity towards a polymerization reaction create strong Ti–N bonds in the composite, and allow for charge carrier formation, which is important for degradation of organic contaminants [57]. The order of photodegradation ability of bisphenol A (BPA) is g-C₃N₄-Melamine in N₂ atmosphere ≈ g-C₃N₄-dicyandiamide > g-C₃N₄-dicyandiamide in N₂ atmosphere > g-C₃N₄-Melamine > g-C₃N₄-Urea ≈ g-C₃N₄-Urea in N₂ atmosphere [58]. The differences between the catalytic activities are mainly due to the different preparation procedures, which can change the type, the density of active sites, the network of sp² hybridized carbon, nitrogen, and oxygen-containing functional groups [58]. Jung et al. showed that the precursor could affect the morphology of the g-C₃N₄-based systems [59]. The good binding of dicyandiamide (DCDA) to ZnO nanoparticles, lead to the formation of the core-shell morphology DCDA-CNZ composite, resulting in improving the degradation of methylene blue by charge transfer [59]. In contrast, due to the weak interaction of thio and urea with ZnO, thio and urea-CNZ have a porous and segregated morphology and produce gases during the polymerization [59].

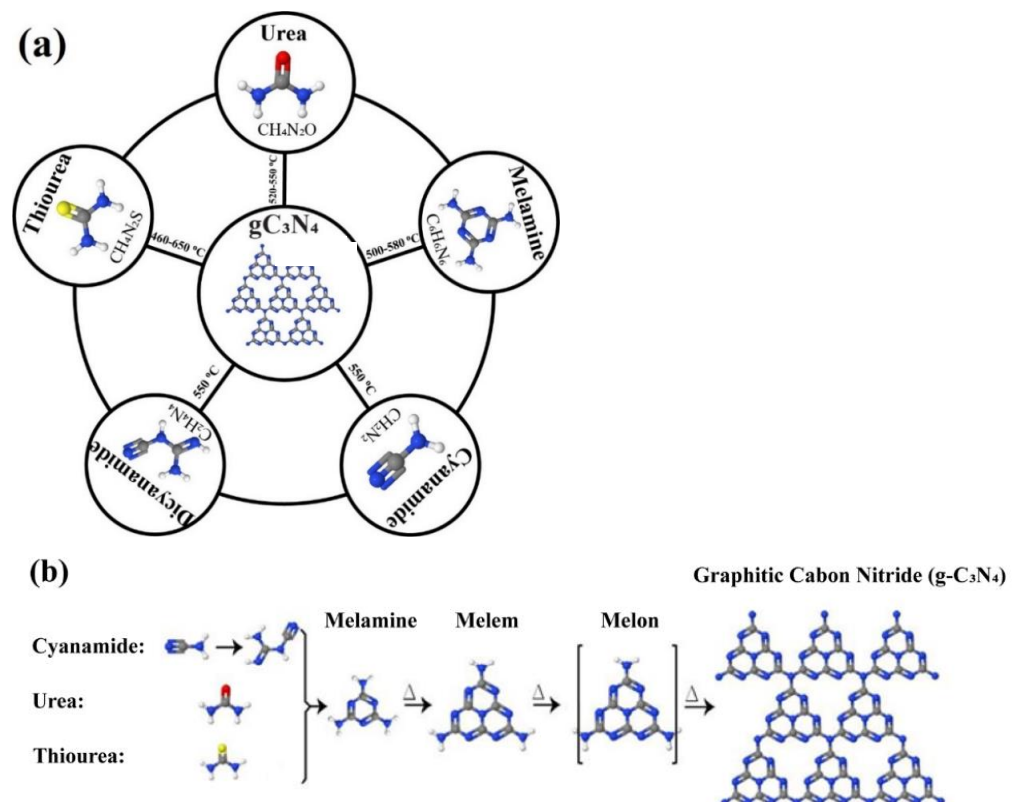


Figure 6. (a) Synthesis precursors and calcination temperature for g-C₃N₄ preparation; (b) synthesis procedure using cyanamide (dicyanamide), urea, thiourea, and melamine for g-C₃N₄ synthesis (gray, blue, red, yellow, and white balls are carbon, nitrogen, oxygen, sulfur, and hydrogen atoms, respectively).

As shown in Figure 6b, melamine can form from urea, thiourea, and cyanamide (dicyanamide), and then converted to tri-s-triazine (and melam) rings at ~ 335 and ~ 390 °C, respectively, and subsequently, g-C₃N₄ discovered from tri-s-triazine polymerization, by heating to 520 °C. Besides, the skeleton becomes prepared over 600 °C, and then beyond 700 °C, g-C₃N₄ is entirely decomposed into small molecules (e.g., NH₃). Figure 7a,b shows the urea and thiourea conversion mechanism into melamine. In this reaction, oxygen atoms in the urea structure help to facilitate g-C₃N₄ condensation and improve stability [60]. CO₂ and NH₃ are the by-products of the reaction of the melamine formation from urea, which can be recycled again to urea. In the formation of the g-C₃N₄, the presence of a crucible lid is so important since not only it prevents the gasses from escaping, but it also provides high pressure in the synthesis atmosphere, which is necessary for the preparation of g-C₃N₄. In other words, a covered alumina crucible should be used during thermal analysis to avoid melamine sublimation.

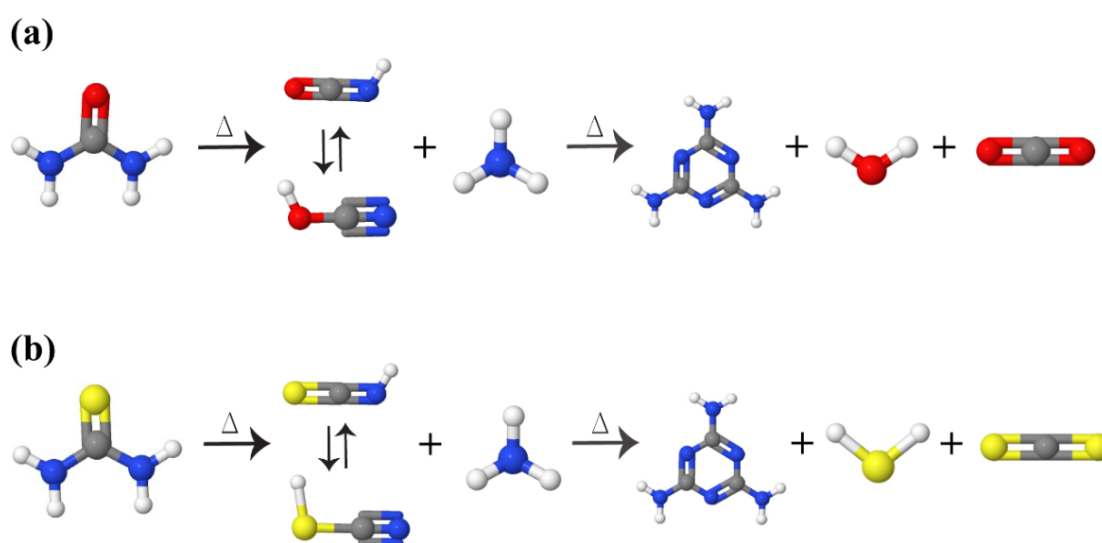


Figure 7. Polymerization of (a) urea and (b) thiourea into a g-C₃N₄ at high temperatures (gray, blue, red, yellow, and white balls are carbon, nitrogen, oxygen, sulfur, and hydrogen atoms, respectively).

During the heating reaction, the created gas bubbles act as soft templates to produce 24 nm pores in the yellow-colored graphitic carbon nitride [61]. Thiourea is used as another precursor for the g-C₃N₄ formation, and sulfur content improves the connectivity and packing of g-C₃N₄ sheets [60]. The overall conversion of urea and thiourea to melamine is endothermic. The first endothermic reaction of changing urea and thiourea to melamine is at temperatures higher than their melting temperatures, which are ~ 133 and ~ 180 °C, respectively, while at the higher temperature, melamine and heptazine were prepared in low pressure at, e.g., atmospheric pressure. To be more specific, the second reaction requires preheating to 260–280 °C to decompose urea in the presence of ammonia, passed over the activated alumina, silica gel, silica-alumina gel, or alumina gel. In order to completely form melamine, the vapors obtained from the first reaction should be maintained at ~ 400 °C [62]. It should also be noted that the g-C₃N₄ can be prepared by cyanamide and dicyanamide. Specifically, by polycondensation of cyanamide molecules and dicyandiamide, melamine was prepared at ~ 203 and ~ 234 °C, respectively [2].

5. g-C₃N₄ Modifications

5.1. Doping

The presence of heptazine ring in the g-C₃N₄ affects electronic structure, toxicity, and density, and is important for applications, especially as biosensors, for photocatalytic hydrogen evolution, and CO₂ conversion. The density-functional theory (DFT) showed that the bandgap of the fully condensed g-C₃N₄ is lower than melam, and polymeric melon, which

are 2.1 eV, 3.5 eV, and 2.6 eV, respectively [63–65]. As previously reported, the polymeric melon has a bandgap close to the defect containing bulk $g\text{-C}_3\text{N}_4$ [64]. There are mainly two types of experimental results to calculate the semiconductor bandgap. Specifically, the position of the conductive band (CB) and valance band (VB) can be measured by electrochemical impedance spectra (EIS), and Mott–Schottky (M–S) curve [63]. The result of both methods showed that the position of the conduction band (CB) and valance band (VB) of the $g\text{-C}_3\text{N}_4$ is about -1.3 eV and 1.4 eV, respectively [3,66]. The semiconductors' band edges can be tuned by functionalizing, doping, compositing with other materials. Liu et al. reviewed element-doped carbonized nitrogen in detail and investigated their organic pollutants degradation applications [3]. Ai et al. showed that the bandgap value of the phosphate doped $g\text{-C}_3\text{N}_4$ decreased from 2.57 eV to 2.49 eV, 2.43 eV, and 2.41 eV by increasing the content of the P element [67]. Other researchers demonstrated that the higher O, Na, Ag, and Co content doped to the $g\text{-C}_3\text{N}_4$ structure leads to the lower bandgap value [68–71]. Li et al. investigated the effect of different ratios of Sm to $g\text{-C}_3\text{N}_4$. They illustrated that by varying the percentile molar ratios of $\text{Sm}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ with melamine from zero to 0.01%, 0.025% and 0.05%, the bandgap decreased from 2.63 eV to 2.57 eV, 2.50 eV, and 2.44 eV, respectively, so Sm narrowed the $g\text{-C}_3\text{N}_4$ bandgap [72]. Table 1 is illustrated the effect of different dopants on the band edge position. We will further discuss and analyze the properties and applications of different $g\text{-C}_3\text{N}_4$ –metal oxides. $g\text{-C}_3\text{N}_4$ is a metal-free semiconductor, which possesses a narrow bandgap suited for visible light absorption (45% of solar energy output) [66]. To have an in-depth investigation of the optical properties of the $g\text{-C}_3\text{N}_4$, we will investigate some characteristics of the synthesized $g\text{-C}_3\text{N}_4$ such as photoluminescence (PL) and UV-vis spectra. The origin and nature of the PL emission come from three different transition pathway including $\pi^*-\pi$, σ^* —the nitride atom bridge's lone pair (LP), and the π^* –LP transition.

5.2. Metal Oxide-Based $g\text{-C}_3\text{N}_4$ Nanocomposite

Different types of metal oxides, such as TiO_2 , ZnO , WO_3 , iron oxide, tin oxide, etc., can improve the photocatalytic efficiency of the $g\text{-C}_3\text{N}_4$ by reducing the electrons-holes recombination and promoting the charge carriers' separation. Consequently, metal oxide-based $g\text{-C}_3\text{N}_4$ nanocomposites can be used in different applications with enhanced electric, magnetic, and photocatalytic properties, such as H_2 generation, CO_2 reduction, NO oxidation, degradation of organic and inorganic dyes and other organic material, removal of toxic metal species, especially Cr (VI) from water, antibodies decontamination, solar cells, sensing, etc. [100–104]. In this part of the review, some metal oxide-based $g\text{-C}_3\text{N}_4$ heterojunction structures are compared with the $g\text{-C}_3\text{N}_4$.

There are five types of charge carrier separation for $g\text{-C}_3\text{N}_4$ –metal oxide photocatalysts:

- (1) Type I heterojunction,
- (2) Type II heterojunction,
- (3) Z-scheme heterojunction,
- (4) p-n heterojunction,
- (5) Schottky junction.

Most $g\text{-C}_3\text{N}_4$ –metal oxide photocatalysts show type II and Z-scheme mechanisms for charge carrier separation. In this section, we will discuss these two heterojunction types.

In type II heterojunctions, two semiconductors are bound to form a stable heterojunction, and the position of the VB of semiconductor A is higher than that of semiconductor B. In this case, because of the difference in voltages, the photoinduced hole migrated from the VB of semiconductor B to that of semiconductor A (Figure 8a). On the other side, electrons are transferred from CB of semiconductor A to that of semiconductor B. The enhanced electrons and holes separation will reduce the rate of the recombination and promote the electrons' lifetime. The construction of type II systems is highly desired for photocatalysis for different applications [7].

Table 1. The electrical properties and application of doped g-C₃N₄.

Doping Element	E _c	E _g	E _v	Application	Further Explanation	Ref
Phosphorus	−1.11 eV	2.55 eV	1.44 eV	Catalytic aromatic alcohols	The presence of P enhances the aldehyde selectivity.	[73]
	−0.33 eV	2.58 eV	2.25 eV	Photocatalytic hydrogen evolution	The g-C ₃ N ₄ tube doped with P improves light absorption. The quantum efficiency of the P-doped g-C ₃ N ₄ tube is 4.7 and 22.4 times higher than that of the g-C ₃ N ₄ tube and bulk g-C ₃ N ₄ .	[74]
	−1.34 eV	2.79 eV	1.44 eV	Photocatalytic CO ₂ conversion	The P-modified g-C ₃ N ₄ demonstrates the highest photocatalytic efficiency.	[75]
	−1.17 eV	2.69 eV	1.52 eV	Photocatalytic hydrogen evolution	The P-doped structure has a high efficiency in accordance with the recombination, migration, and separation of electron-hole pairs.	[76]
Sulfur	−1.04 eV	2.92 eV	1.88 eV	Photocatalytic nitrogen fixation	Sulfur enhances the adsorption and activation of N ₂ molecules of g-C ₃ N ₄ porous nanosheets and uses for photocatalytic nitrogen fixation.	[77]
	−1.23 eV	2.80 eV	1.57 eV	Photocatalytic hydrogen evolution	The H ₂ generation rate of N-doped MoS ₂ and S-doped g-C ₃ N ₄ is about 23 and 38 times higher than that of pure SCN and NMS with 28.8 μmol/g/h and 17.4 μmol/g/h, respectively.	[78]
	−1.3 eV	2.67 eV	1.34 eV	Photocatalytic hydrogen evolution	The BiPO ₄ /S-C ₃ N ₄ improves photocatalytic activity by facilitating carrier transportation.	[79]
	−1.3 eV	2.69 eV	1.39 eV	Photocatalytic bisphenol degradation	Ag-S-C ₃ N ₄ enhanced the photocatalytic activity since Ag has a great electron storage ability.	[80]
Oxygen	−1.32 eV	2.66 eV	1.34 eV	Photocatalytic hydrogen evolution	Sulfur promotes the photocatalytic ability of hydrogen evolution about four times higher than the bulk g-C ₃ N ₄ .	[81]
	−0.88 eV	2.61 eV	1.73 eV	Photocatalytic CO ₂ reduction	The porous O-doped graphitic carbon nitride reveals enhanced photocatalytic activity.	[82]
	−0.76 eV	2.57 eV	1.84 eV	Photocatalytic hydrogen evolution	This result of the band edge value is related to the 1.1% oxygen content mass percentage.	[83]
	−0.37 eV	2.53 eV	2.15 eV	Photocatalytic hydrogen evolution	This result of the band edge value is related to the 2.3% oxygen content mass percentage.	[83]
	−1.08 eV	2.93 eV	1.85 eV	Photocatalytic hydrogen evolution and 2,4-dinitrophenol	The oxygen dopant with Pt exhibits excellent photocatalytic hydrogen evolution in overall water splitting with 29.6 μmol/(g·h), and O-g-C ₃ N ₄ NR reached up to approximately 100% removal efficiency of 2,4-dinitrophenol within 75 min.	[84]
	1.51 eV	2.70 eV	−1.19 eV	photocatalytic water splitting	This band edge value is related to CN-x = 0 (x refers to the quantity of citric acid (gr)).	[85]
1.50 eV	2.62 eV	−1.16 eV	This band edge value is related to CN-0.2.			
1.46 eV	2.52 eV	−1.06 eV	This band edge value is related to CN-0.4.			
Carbon	1.46 eV	2.49 eV	−1.03 eV	Thermal oxidation etching process	This band edge value is related to CN-0.6.	[86]
	−1.13 eV	2.54 eV	1.41 eV		C-doped g-C ₃ N ₄ improves the catalytic activity by extending the visible light absorption.	
Boron	−0.8 eV	2.8 eV	2 eV	Photocatalytic Oxygen evolution, Cr(VI) reduction	This structure is used for Cr(VI) reduction and O ₂ generation simultaneously.	[87]
Nitrogen	−0.5 eV	2.4 eV	1.9 eV	Photocatalytic tetracycline degradation	This band edge value is related to the nitrogen-doped g-C ₃ N ₄ used for Photocatalytic tetracycline degradation.	[88]
	−0.6 eV	2.5 eV	1.9 eV	Photocatalytic phenol degradation	This band edge value is related to the nitrogen-doped g-C ₃ N ₄ nanosheets.	[89]
	−0.33 eV	1.82 eV	1.49 eV	N-doped g-C ₃ N ₄ possesses a narrow bandgap since the N atom introduced an inter-bandgap, resulting in the redshift in the absorption UV-vis peak.		

Table 1. Cont.

Doping Element	E_c	E_g	E_v	Application	Further Explanation	Ref	
Metal	Na	−1.16 eV	2.77 eV	1.36 eV	17 α -ethynylestradiol mineralization	The photostable Na doped g-C ₃ N ₄ content causes the photoabsorption enhancement.	[70]
	K	−1.08 eV	2.72 eV	1.64 eV	Photocatalytic CO ₂ reduction	K content causes defects leading to improving catalytic activity by reducing the electron-hole recombination.	[90]
	Ti	−1.02 eV	2.5 eV	1.48 eV	Photocatalytic enhancement	Ti-doped g-C ₃ N ₄ caused narrower bandgap and reduced carrier recombination resulting in higher absorption.	[91]
	Mn	−0.59 eV	2.56 eV	1.97 eV	Photocatalytic methylene blue degradation	The Mn-doped g-C ₃ N ₄ nanoribbon reveals a great potential photocatalytic agent for water splitting coupling with MB degradation.	[92]
	Ag	-	2.60 eV	-	Photocatalytic oxidation of methylene blue	The higher Ag content leads to the lower bandgap of the structure, and a lower recombination rate is observed in the Ag-doped g-C ₃ N ₄ .	[93]
	Fe	−1.10 eV	2.50 eV	1.40 eV	Environmental pollution control	Fe ³⁺ with nitrogen in heptazine forms a σ - π bond and can accelerate the electron-hole separation.	[94]
Co-doped	Co	−0.36 eV	2.62 eV	2.26 eV	Photo-electrochemical water oxidation	Co-doped g-C ₃ N ₄ reduces the electron-hole recombination rate and demonstrates promising photocurrent and electrical conductivity.	[95]
	P, O	−0.80 eV	2.30 eV	1.50 eV	Photocatalytic fluoroquinolone antibiotics degradation	The degradation rate of enrofloxacin was 6.2 times higher for phosphorus and oxygen co-doped graphitic carbon nitride (POCN) than g-C ₃ N ₄ .	[96]
	P, S	-	2.6 eV	-	Photocatalytic hydrogen evolution	The high photocatalytic activity can be observed because of the synergic impact of P and S co-doping.	[97]
	B, F	-	2.72 eV	-	Photocatalytic hydrogen evolution	B, F co-doped g-C ₃ N ₄ improves the charge generation and the separation efficiency.	[98]
	Na, O	-	2.72 eV	-	Photocatalytic hydrogen evolution	Na, O co-doped g-C ₃ N ₄ reveals that photocatalytic H ₂ production activity was seven-fold improved by enhancing absorption of UV-vis spectra.	[99]

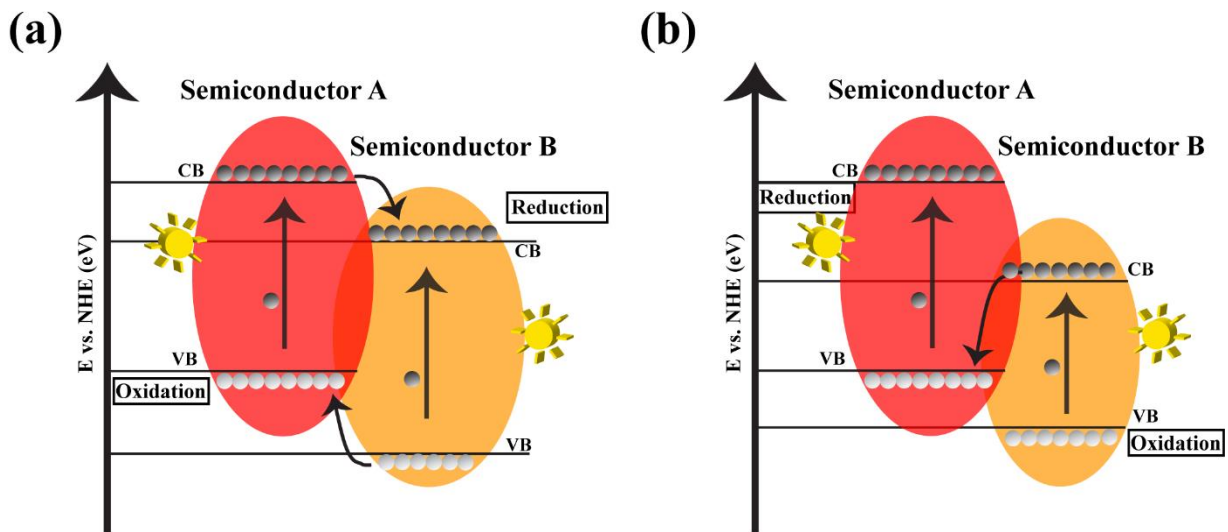


Figure 8. The two most common heterojunction types in $g\text{-C}_3\text{N}_4$ –metal oxide photocatalysts: (a) type II heterojunction, (b) Z-scheme heterojunction (white and dark balls are holes and electrons, respectively).

The other heterojunction type is the direct Z-scheme photocatalytic system, which was initially suggested by Bard et al. in 1995 [105]. As shown in Figure 8b, in this heterojunction type, the generated electrons on the CB of semiconductor B transfer to the VB of semiconductor A and combine with the photogenerated holes. This type of photocatalysts can be helpful for both reducing the recombination by an increase of the electrons and holes separation and improving the redox ability [7]. Even if the electrons, and holes combine and generate $h\nu$ in these heterojunctions, other photogenerated carriers can replace them.

Different patterns of the migration of electrons and holes are due to the driving force of the electric field, formed by the band edge positions of different semiconductors. Depending on the electrical field direction, charge carriers start moving to reduce the energy of the system. Thus, different systems are defined based on the migration of electrons and holes in the heterojunction. The difference in each type is mainly due to the movement of electrons and holes. All types can be used in different photocatalytic activities.

5.2.1. TiO_2 - $g\text{-C}_3\text{N}_4$

Among the investigated semiconductor photocatalysts, TiO_2 has a suitable conduction band position, excellent stability, cost-effective preparation approach and is one of the most promising catalytic materials [106]. Fujishima and Honda were pioneers who researched using TiO_2 photocatalytic behavior in 1972 [107]. To improve TiO_2 's photocatalytic efficiency, researchers would like to reduce the bandgap of the system by doping with other elements and compositing with other compounds to absorb visible light energy [108]. To deal with TiO_2 's limitations, researchers have been using doping elements and compositing with organic material, such as conjugated polymers and $g\text{-C}_3\text{N}_4$, since they have a narrow bandgap [108]. Boron is an effective dopant, which can improve the photocatalytic applications of TiO_2 coupled with carbon nitride, as shown by Christoforidis and coworkers [109]. In another study conducted by this research group, TiO_2 and carbon nitride nanosheets were synthesized by hydrothermal in-situ approach, improving the catalytic application. The mentioned materials have high porosity to ensure a high concentration of reactants in the vicinity of catalytic sites, used for CO_2 reduction [110].

Various synthetic methods such as co-calcination, hydrothermal treatment, solvothermal, and microwave-assisted to prepare the $g\text{-C}_3\text{N}_4$ - TiO_2 heterojunction [111–113]. The preparation of $g\text{-C}_3\text{N}_4$ - TiO_2 heterojunction widely includes the hydrothermal and calcination method [114]. Figure 9 illustrates the schematic of the TiO_2 / $g\text{-C}_3\text{N}_4$ preparation route.

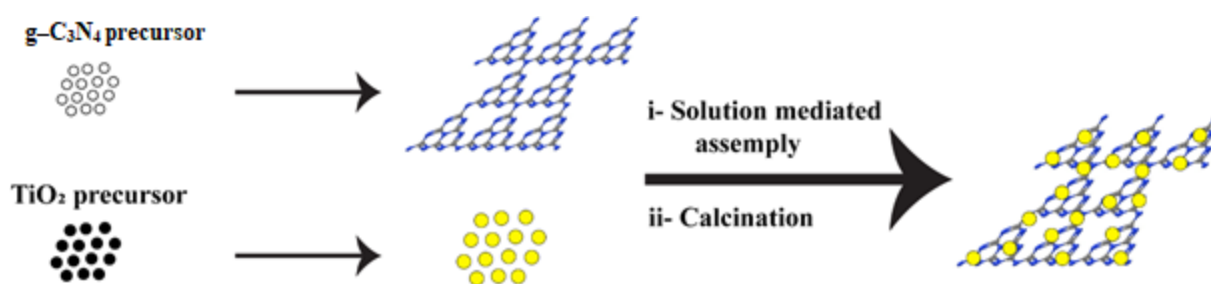


Figure 9. Schematic illustration of $\text{TiO}_2/\text{g-C}_3\text{N}_4$ heterojunctions synthesis approach.

The conductive band of $\text{g-C}_3\text{N}_4$ ($E_{\text{CB}} = -1.4$ eV) is higher than those of anatase TiO_2 ($E_{\text{CB}} = -0.5$ eV), so the proper bandgap alignment helps to reduce the electron-hole recombination by increasing their separation and boost the space charge accumulation at the interface. Recombination is defined as a process of electrons and holes annihilation. The most common type of recombination is known as radiative recombination, which occurs when electrons and holes in a conductive and valence band, respectively, recombine and emit a photon. As discussed in the following sections, the presence of electrons and holes will assist us in different stages of photocatalytic activities. So, suppressing the charge recombination is crucial in the photocatalytic applications of semiconductors and heterojunctions. At the end of section three, we talked about how $\text{g-C}_3\text{N}_4$ -based heterojunction separates the position of the electrons and holes. The charge carrier separation makes electron-holes recombination less favorable, so we can use these heterojunctions for photocatalytic activities.

Figure 10a illustrates the conduction, valence, and bandgap position of the $\text{g-C}_3\text{N}_4$ and TiO_2 and reveals that the $\text{g-C}_3\text{N}_4$ - TiO_2 structure improves the photo-induced electrons flow from the $\text{g-C}_3\text{N}_4$ conductive band to that of TiO_2 , which promotes the photoelectrical ability of the final composite. Thus, the photogenerated electrons tend to accumulate in the TiO_2 conductive band since the conductive band of TiO_2 is more positive than that of $\text{g-C}_3\text{N}_4$, (Figure 10a). In contrast, the holes transfer in an inverse way, which can provide type II heterojunction [112,115]. Due to the high recombination barrier, the heterojunction provides a high interfacial area for facilitating carrier transformation and separation to suppress the electron-hole recombination to improve the photocatalytic activity.

Kočí et al. successfully deposited TiO_2 on the $\text{g-C}_3\text{N}_4$ surface by hydrothermal approach followed by calcination processing [116]. In this research, the mixture of TiO_2 and $\text{g-C}_3\text{N}_4$, prepared by thermal hydrolysis and polycondensation from melamine, respectively, should be mixed in distilled water for 16 h in an air atmosphere dried at 60 °C. Finally, the dried sample should be maintained at 450 °C in a covered crucible with a heating ramp of 15 °C/min in the air in a muffle furnace. The highest photocatalytic performance was obtained under UVA ($\lambda = 365$ nm) irradiation compared to other types of ultraviolet (UV) rays (UVB, and UVC), which is attributed to the high separated charge carrier. The low rate of recombination leads to the great photocurrent stability. In another work on $\text{g-C}_3\text{N}_4$ - TiO_2 heterojunction structure, Alcludia-Ramos et al. demonstrated that the prepared heterojunction has a higher photocatalytic efficiency than the individual $\text{g-C}_3\text{N}_4$ and TiO_2 [112]. The researcher also showed that solvothermal synthesis enhances tri-s-triazine's carbonization [117]. Miranda et al. used an impregnation method to prepare $\text{g-C}_3\text{N}_4$ - TiO_2 [118]. The presence of the $\text{g-C}_3\text{N}_4$ in the heterojunction endows the high specific area to the structure. The researchers also used the photochemical reduction method to prepared $\text{g-C}_3\text{N}_4$ - TiO_2 based nanocomposite. They also demonstrated that in the $\text{TiO}_2/\text{g-C}_3\text{N}_4/\text{G}$ composite, TiO_2 was a semiconductor to capture visible light and also prevents $\text{g-C}_3\text{N}_4/\text{G}$ stacking [119].

Several parameters affect the $\text{g-C}_3\text{N}_4$ - TiO_2 heterojunction structure. Various research works have been investigated the effect of the $\text{g-C}_3\text{N}_4$'s ratio. Wang et al. prepared this microstructure with 10%, 30%, 50%, 70% of $\text{g-C}_3\text{N}_4$, which are labeled with ($x = 10, 30,$

50, and 70) [110]. All the XRD results from different publications have shown the same diffraction pattern. As a case in point, the XRD pattern of $g\text{-C}_3\text{N}_4\text{-TiO}_2$ is illustrated in Figure 10b. In the TiO_2 diffraction pattern, the peaks at 25.5, 37.7, 48.2 and 54.1 °C are related to the (101), (004), (200) and (105) planes, respectively. Furthermore, there is no $g\text{-C}_3\text{N}_4$ characteristic peak at 13.1 °C in the $g\text{-C}_3\text{N}_4\text{-TiO}_2$ hybrid sample, which may be attributed to the low crystallinity of $g\text{-C}_3\text{N}_4$ compared to the TiO_2 in the microstructure. However, the peak at 27.5 is obviously observed in all samples. The more $g\text{-C}_3\text{N}_4$ ratio in the structure leads to the higher XRD intensity at 27.5 °C and the lower peak intensity at 25.5 °C [120]. TiO_2 possesses a high crystallinity so the characteristic peak at 25.5 °C is higher than 27.5 °C. There are no obvious changes in other characteristic peaks in the composite XRD patterns. The FT-IR spectra of the $g\text{-C}_3\text{N}_4\text{-TiO}_2$ structures are demonstrated in Figure 10c [120]. In all FT-IR analyses, stretching vibration of the Ti–O and Ti–O–Ti is observed at about 475 cm^{-1} . Additionally, peaks at 3419 cm^{-1} are attributed to the adsorbed moisture and hydroxyl group in the structure. The peak at 2360 cm^{-1} that appeared in all samples is mainly due to the adsorbed CO_2 . All the characteristic peaks at TiO_2 and $g\text{-C}_3\text{N}_4$ are observed in the composite samples. The pore size distribution of the $g\text{-C}_3\text{N}_4$, TiO_2 , $g\text{-C}_3\text{N}_4\text{-TiO}_2$ is illustrated in Figure 10d [120]. The pore size at ~3.5 nm, 10 nm, and 3.8 nm, 32 nm is due to the presence of TiO_2 , and $g\text{-C}_3\text{N}_4$, respectively. Additionally, the BET specific area of the $g\text{-C}_3\text{N}_4\text{-TiO}_2$ hybrid structure was increased at the higher $g\text{-C}_3\text{N}_4$ content. Finally, the higher the bandgap of TiO_2 , the lesser the absorption wavelength at higher than 400 nm (Figure 10e). The $g\text{-C}_3\text{N}_4$ revealed the absorption band extending to about 430 nm, which is mainly due to the low bandgap [120]. The blue shift in the absorption spectra was also demonstrated at a higher $g\text{-C}_3\text{N}_4$ ratio in the structure. It was shown bandgaps calculation of the $g\text{-C}_3\text{N}_4$, TiO_2 , $g\text{-C}_3\text{N}_4\text{-TiO}_2$. The results revealed that the bandgap of CNT50 samples is 2.92 eV, which is like the bulk $g\text{-C}_3\text{N}_4$ with the bandgap of 2.9 eV and narrower than the bandgap of TiO_2 (3.20 eV). Due to the narrow bandgap and heterojunction formation, the hybrid composite not only suggested the higher generation of the electron and hole but also improved photocatalytic activity [120,121]. Li et al. synthesized $g\text{-C}_3\text{N}_4\text{@TiO}_2$ hollow sphere nanostructure with high crystallinity [122]. In this work, researchers used the different ratios of TiO_2 hollow sphere and melamine (1:2, 1:4, and 1:8) in the solution, which are called HS-CNTO1, HS-CNTO2, and HS-CNTO3, respectively. They demonstrated that the recombination rate of the photogenerated electron-holes decreased by introducing the TiO_2 in the $g\text{-C}_3\text{N}_4$ structure. In all samples, the sharp PL emission peak at 455 nm can be observed. The intensity of the PL peak reduced as the $g\text{-C}_3\text{N}_4$ content ratio decreased [122]. In addition, as can be seen in Figure 10f, the electron resistance decreased when $g\text{-C}_3\text{N}_4\text{-TiO}_2$ heterojunction was used. Additionally, stability is another vitally important factor in determining the photocatalytic activities, and it is shown that the $g\text{-C}_3\text{N}_4\text{-TiO}_2$ heterojunction demonstrates excellent stability after three to five cycles under different circumstances [117,120,122,123].

Many factors may affect the $g\text{-C}_3\text{N}_4\text{-TiO}_2$ heterojunction productivity and improve the photocatalytic activity by enhancing the charge carrier separation and prevent recombination [124–126]. Rathi and coworkers showed that the $\text{CuNi@g-C}_3\text{N}_4\text{-TiO}_2$ nanocatalyst had a 3-fold and 5-fold higher photocatalytic activity than bare $g\text{-C}_3\text{N}_4$ and TiO_2 nanorod for Rhodamine B degradation. Besides, the photocurrent density of the TiO_2 nanorod, bare $g\text{-C}_3\text{N}_4$, $\text{Cu@g-C}_3\text{N}_4$, $\text{Ni@g-C}_3\text{N}_4$, and $\text{TiO}_2/\text{CuNi@g-C}_3\text{N}_4$ is 0.108 mA/cm^2 , 0.377 mA/cm^2 , 0.530 mA/cm^2 , 0.6012 mA/cm^2 , and 0.890 mA/cm^2 , respectively. It should also be mentioned that the charge separation was promoted since the presence of Cu and Ni species [126]. In another work, researchers also prepared $\text{Ti}^{3+}\text{-TiO}_2/\text{O-g-C}_3\text{N}_4$ heterojunctions via a hydrothermal approach [125]. In this synthesis approach, 1 g $g\text{-C}_3\text{N}_4$ should disperse with titanium oxohydrides sol precursor at room temperature for 20 min, ultrasonically. The collected sample should transfer into the Teflon-lined autoclave at 160 °C for 27 h and then be washed and dried at 60 °C for 3 h. This method was used to prevent the $g\text{-C}_3\text{N}_4$ aggregation and fabricate exfoliated $g\text{-C}_3\text{N}_4$ nanosheets. The synthesized heterojunction significantly decreased the regenerated electron-hole pairs' recombination. Additionally,

the conductivity is greatly enhanced and widens the light absorption range by adding the Ti^{3+} , and O. P is another element used to improve the heterojunction connection and promote photocatalytic activity by facilitating the carriers' transfer and separation [127,128].

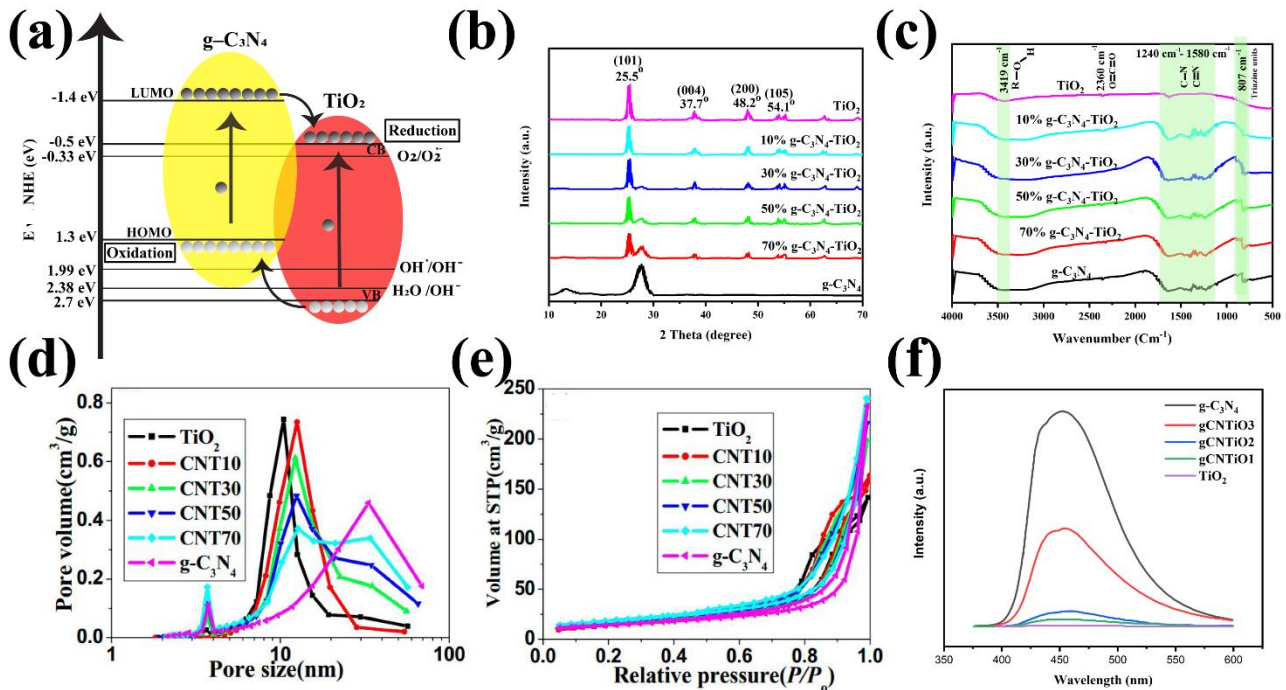


Figure 10. (a) Proposed mechanism for charge transfer of type II of the $\text{g-C}_3\text{N}_4\text{-TiO}_2$ heterojunction interface under visible light irradiation; (b) XRD patterns; (c) FT-IR spectra of the $\text{g-C}_3\text{N}_4\text{-TiO}_2$ heterojunction; (d) the pore size distribution curves; (e) UV-vis spectra of the prepared TiO_2 , $\text{g-C}_3\text{N}_4$, and $\text{TiO}_2\text{-g-C}_3\text{N}_4$ composites, Copyright © 2022 Elsevier [120]; (f) PL emission spectrum of TiO_2 , $\text{g-C}_3\text{N}_4\text{-TiO}_2$, and $\text{g-C}_3\text{N}_4$ products, Copyright 2019 © American Chemical Society [122].

Various novel metal nanoparticles are leading to the improvement of photoexcited semiconductors because the surface plasmonic effect is beneficial to reduce the photogenerated electron-hole recombination, improve the efficiency of visible light absorption and photocatalytic activity. Silver, and gold nanoparticles, for instance, have high stability and good conductivity. Au nanoparticles can increase the electron concentration onto their surface and enhance and extended adsorption for catalytic activity by its surface π bond [129–131]. Ag nanoparticles (AgNPs) are also used to modify the $\text{g-C}_3\text{N}_4\text{-TiO}_2$ heterojunction [132–135]. The presence of AgNPs not only promotes the visible light response due to the surface plasmon resonance (SPR) effect, but also, they can result in capturing the electrons, to separate them, and transfer them more easily. These electrons react with the absorbed O_2 on the AgNPs modified $\text{TiO}_2\text{@g-C}_3\text{N}_4$ to form O_2^- . With reference to the high specific area, electrical conductivity, and mobility, different graphene-based materials such as reduced graphene oxide [119,136]. Other materials can also be used to modified $\text{g-C}_3\text{N}_4\text{-TiO}_2$ heterojunction to increase the specific area and separate the photo-induced electron-hole pairs to enhance photocatalytic efficiency [137–139].

The structure and morphological evaluation of the prepared $\text{g-C}_3\text{N}_4\text{-metal oxide}$ -based heterojunctions have been investigated in different research works [140–143]. Jo et al. revealed structural properties of $\text{g-C}_3\text{N}_3\text{-TiO}_2$ heterojunctions, which are consistent with other similar composites [144]. Figure 11a showed the transmission electron microscope (TEM) micrograph and the selected area electron diffraction (SAED) pattern of 5%- $\text{g-C}_3\text{N}_4/\text{TiO}_2$ nanoparticles. Researchers observed that $\text{g-C}_3\text{N}_4$ nanolayer uniformly is covered with TiO_2 nanoparticles, which suggested an intimate interface between them. Besides, the inset Figure 11a showed that the circular rings corresponded to the (101), (004),

(200), and (105) planes of the polycrystalline TiO_2 nanoparticles anatase phase. A TEM image showed that the TiO_2 nanoparticles are deposited onto the layered structure on $g\text{-C}_3\text{N}_4$. Moreover, the high-resolution transmission electron microscope (HR-TEM) images of the 5%- $g\text{-C}_3\text{N}_4/\text{TiO}_2$ nanotube indicates the interplanar distance of 0.350 nm, attributed to the (101) plane of anatase phase of TiO_2 (Figure 11b,c). In addition, elemental mapping analysis of the 10%-CN/TNP (Figure 11d) suggested that TiO_2 is uniformly present onto the $g\text{-C}_3\text{N}_4$ surface. Figure 11d also revealed the presence of Ti, O, C, and N, which suggested the co-existence of both layered $g\text{-C}_3\text{N}_4$ and TiO_2 nanotube [144].

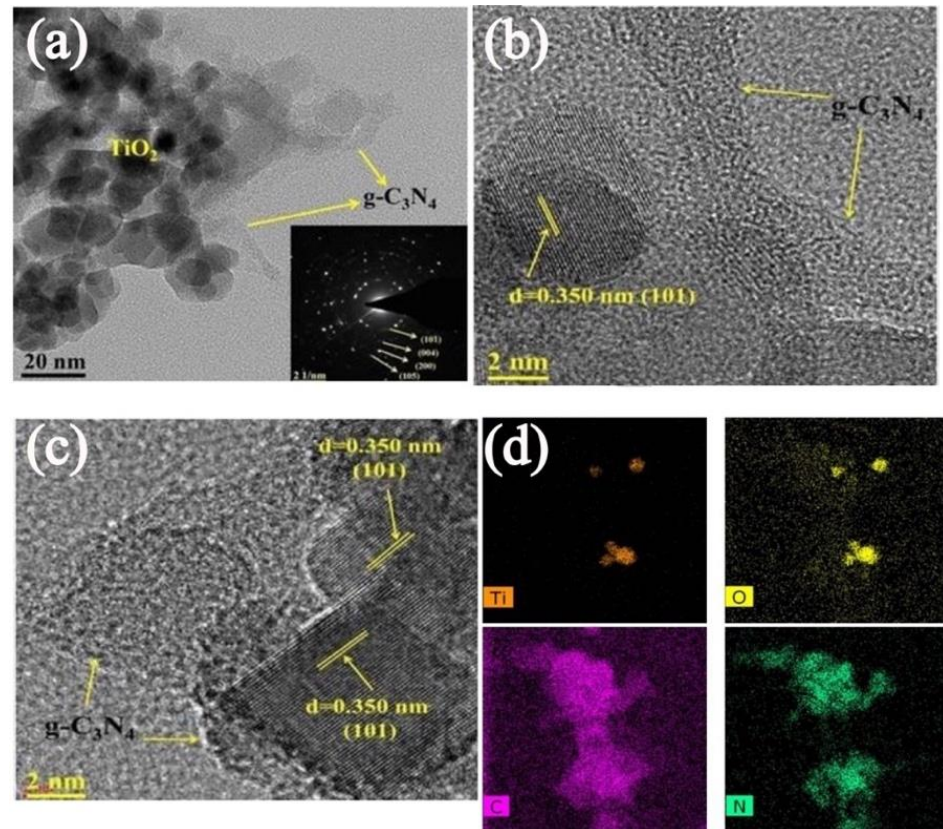


Figure 11. (a) TEM images of 5%- $g\text{-C}_3\text{N}_4/\text{TiO}_2$ nanoparticles (inset SAED pattern); (b,c) HRTEM images of 5%- $g\text{-C}_3\text{N}_4$; (d) elemental mapping image of Ti, O, C, N elements of the 10%- $g\text{-C}_3\text{N}_4/\text{TiO}_2$ composite, Copyright © 2022 Elsevier [144].

5.2.2. ZnO- $g\text{-C}_3\text{N}_4$

ZnO nanostructures such as nanosheets, nanoplates, and nanorods are semiconductors that have been used for preparing of $g\text{-C}_3\text{N}_4$ -based heterojunctions. Like the TiO_2 , the ZnO bandgap is about $\sim 3.2\text{ eV}$ with E_{CB} and E_{VB} of about 2.7 eV and -0.5 eV , respectively [145]. We showed that the ZnO nanostructure's size, shape, and order could be tuned by an interplay of magnetic and gravity forces [146]. We also demonstrated the enhanced microbial detection capability when the synthesis was conducted under these external forces by changing the materials' electrical resistance based on surface interactions. Because of low-cost of the preparation, having a large surface area, high aspect ratio, proper bandgap energy, minimal toxicity, and good stability, ZnO nanostructures have captured considerable researchers' attention [147,148]. However, ZnO suffers from minimal light absorption (5% of the ultraviolet spectrum of the sun energy). Additionally, a high electron-hole recombination rate is another undesirable factor of ZnO for photocatalytic applications.

To deal with these problems the heterojunction of ZnO with $g\text{-C}_3\text{N}_4$ might be an option for different photocatalytic applications because, coupling $g\text{-C}_3\text{N}_4$ with ZnO nanostructures can improve charge migration, separation and prevent electron-hole recombination.

Researchers have reported various methods for preparing the g-C₃N₄-ZnO structure, such as hydrothermal, solvothermal, atomic layer deposition, etc. [149–153]. Jung et al. synthesized g-C₃N₄-ZnO with various thermal treatment and condensation temperatures (T = 350, 400, 450, 500 °C). The BET specific area and ZnO crystallite size of the prepared Z-scheme g-C₃N₄-ZnO structure is decreased by increasing the thermal treatment temperature [154]. Other researchers who designed g-C₃N₄-ZnO via thermal treatment announced that the optimal amount of the g-C₃N₄ content in the composite is 5.0 wt.% [155]. Melamine and ZnCl₂ should be vigorously stirred for 20 min in a 250 mL beaker, then, Na₂CO₃ is added dropwise into the suspension and stirred magnetically for 30 min, and finally, dried at 60 °C for 30 min. The product was placed into a crucible with a cover to prevent from the volatilization of melamine and heated at 500 °C for 2 h at a rate of 10 °C/min. The g-C₃N₄/ZnO photocatalysts were obtained after deamination treatment at 520 °C for 2 h. The effect of different g-C₃N₄ precursors such as dicyandiamide (DCDA), urea, and thiourea on the g-C₃N₄ and ZnO interaction and structural morphology was investigated in another work [156]. The excellent interaction of the DCDA-ZnO results in the perfect Z-scheme charge transfer core-shell structure with the ZnO core and shell of the g-C₃N₄, and the low electron density of the PL emission resulting in promoting the efficiency of the Methylene Blue (MB) photocatalytic degradation. If the interaction between the precursors, such as urea and thiol, and ZnO is weak, the porous, segregated morphology is obtained. Hydrothermal method to prepare g-C₃N₄-ZnO heterojunction [149] not only is a low-cost preparation but also Zhang et al. revealed that it could detect nine pesticide residues in four different samples simultaneously. The solvothermal synthesis method of the preparation of the g-C₃N₄-ZnO modified the TiO₂ nanotube arrays by using the ethylene glycol solution is reported by Mohammadi et al. [150]. Zhang and coworkers synthesized the g-C₃N₄-ZnO heterojunction composite in the metal ion-containing ionic liquid's presence by the solvothermal method [156]. g-C₃N₄ was added to the ZnCl₂ in an ethanol solution, sonicated, and mixed with NaOH. Then, the mixture should be placed in a 25 mL Teflon-sealed autoclave and maintained at 160 °C for 24 h. After washing with distilled water and absolute ethanol, the dried g-C₃N₄-ZnO powder was provided. The strong interaction between g-C₃N₄ and ZnO result in the higher migration of the generated electrons and slower recombination rate was prepared. They revealed that the power conversion of the structure composed by the solvothermal approach compared to the pure TiO₂ nanotube arrays increases from 1.04% to 2.45%. Besides, the uniform type II heterojunction between g-C₃N₄ and ZnO can also be synthesized by atomic layer deposition (ALD) [151]. The mentioned heterojunction composite, synthesized by the ALD method, possessed a stable dispersion of g-C₃N₄ powder in the reactor, which prevented the charge carrier recombination. Mechanochemistry (mechanical milling) is also employed to prepare g-C₃N₄-ZnO composite [157]. This method provided significant photocatalytic stability and enhanced the composite's photocatalytic activity, which was 3-fold higher than the bulk g-C₃N₄ because of the strong interaction with ZnO. Figure 12 illustrates the schematic of the ZnO/g-C₃N₄ preparation method.

Introducing the ZnO to the g-C₃N₄ is proved that this structure can enhance photocatalytic performance such as charge transfer and separation and decrease the photogenerated carriers' recombination (Figure 13a) [159–163]. In recent years, several types of research have been worked on the characterization of these heterojunctions. For example, Wang et al. demonstrated the eight XRD characteristic peaks for the pure Zn in the g-C₃N₄-ZnO sample, shown in Figure 13b [164]. The reduction in the g-C₃N₄ content results in a decrease in the intensity of its two prominent characteristic peaks. Besides, they also revealed the FTIR analysis of the synthesized composites. The g-C₃N₄-ZnO composites' FTIR peaks show the main peaks of the bulk g-C₃N₄, which are shifted to the lower wavenumber, and this is because of the low strength of the characteristic bonds. The FTIR peaks of the g-C₃N₄-ZnO heterojunction are also similar to those of the main peaks of the g-C₃N₄ wavenumber. The indication announces a chemical bond in the heterojunction between g-C₃N₄ and ZnO so that this structure will improve the charge transfer and photocatalytic efficiency. Moreover,

the UV-vis DRS spectra of the g-C₃N₄, ZnO, and g-C₃N₄-ZnO heterojunction with the different g-C₃N₄ ratio is illustrated in Figure 13c [164]. The absorption edge of the g-C₃N₄ is about 460 nm, attributed to the bandgap of 2.69 eV. Besides, the absorption wavelength of the pure ZnO appeared at 396 nm showing the bandgap of 3.13 eV. Finally, the g-C₃N₄-ZnO revealed the redshift for the higher g-C₃N₄ content, which is extended to the visible-light region. The intense visible light absorption is another significant indication of the strong chemical interaction and more electron-hole generation leading to the photocatalytic activity's improvement [164]. The high photocatalytic activity of the mentioned structure is mainly used to degrade of dyes such as malachite green (MG). After the reaction of the photocatalyst with MG, the photocatalytic rate in 0, 5, 10, 15, 20, 25, 30, 35 and 45 min was measured (Figure 13d) [165]. A similar analysis was conducted on other dye types, such as Rhodamine-B (Rh-B), Congo red (Con-R), and Red ink (RI) solution. The results show the degradation efficiency after 45 min in the presence of the g-C₃N₄ was determined ~97.24%, 82.37%, 70.05% and 46.99% for MG, Rh-B, Con-R, and RI's degradation, respectively [165]. Figure 13e demonstrated the photoinduced charge transfer capability of g-C₃N₄-ZnO nanorod arrays. The photocurrent density of the prepared materials was generated and increased under visible light irradiation. However, the photocurrent density decreased as the illumination was stopped. It was also revealed that the g-C₃N₄-ZnO produced the most photocurrent density compared to the bare ZnO and g-C₃N₄, showing excellent charge transfer and separation. The charge transfer resistance is depicted in Figure 13f, shown impedance spectroscopy (EIS) Nyquist plots [160]. The arc radius in EIS spectra corresponds to the resistance of the interface layer at the photocatalyst surface. As observed in Figure 13f, the smaller arc radius is shown for g-C₃N₄-ZnO than that of g-C₃N₄ and ZnO. To be more specific, the smaller arc radius leads to the lower charge transfer resistance, resulting in the improved photogenerated transformation at the interface. PL spectra announce the photocurrent recombination, separation, and migration rate [160]. The ZnO PL emission wavelengths under the excitation wavelength of 350 nm were low-intensity emissions at 445 nm and 600 nm with low intensity. Besides, the emission spectra of the g-C₃N₄ and g-C₃N₄-ZnO heterojunction under the same excitation wavelength are 452 nm and 506 nm, respectively. Moreover, the higher PL intensity of the composite than that of g-C₃N₄ and ZnO reveals a more recombination rate of photogenerated charge carriers via the Z-scheme pathway. One of the most critical factors in photocatalytic efficiency is the catalyst's lifetime. Recyclability of g-C₃N₄-ZnO is obviously illustrated by the negligible loss of the photocatalytic activity after the five cycles resulting in good stability of the g-C₃N₄-ZnO photocatalyst [166–168].

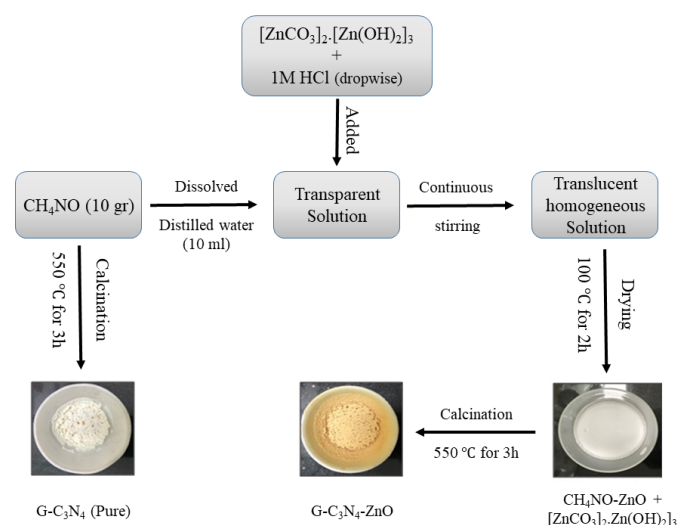


Figure 12. Schematic of the g-C₃N₄-ZnO heterojunction synthesis approach, reproduced from Reference [158].

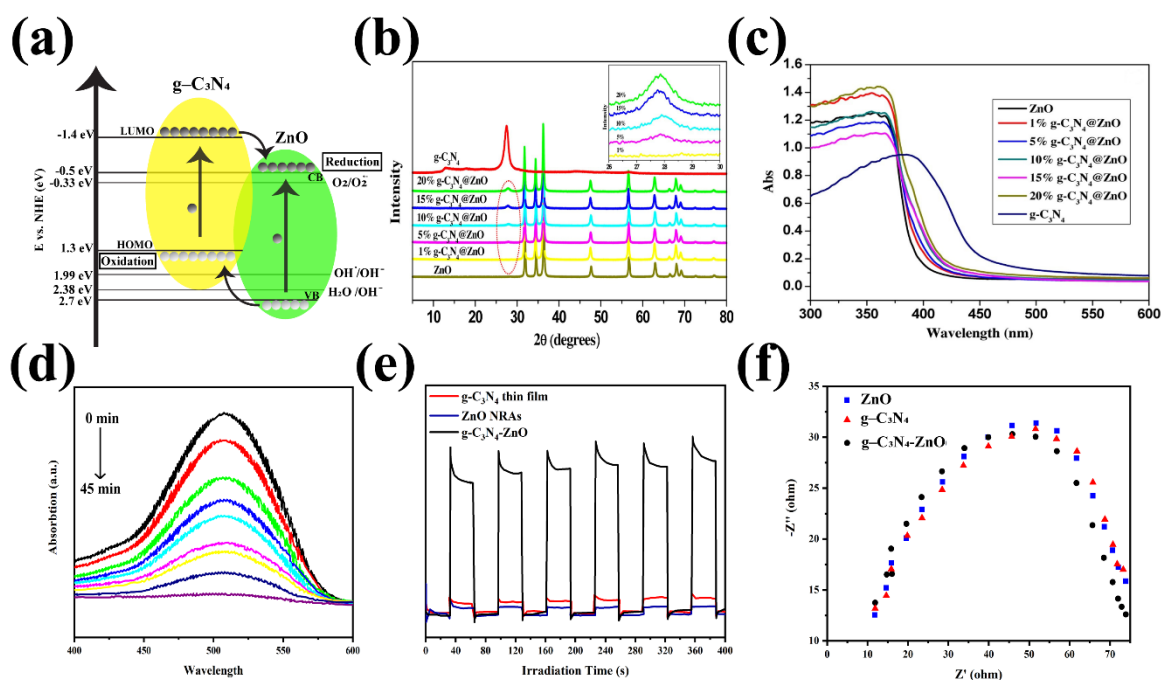


Figure 13. (a) Proposed mechanism for charge transfer of type II of the $g\text{-C}_3\text{N}_4\text{-ZnO}$ heterojunction interface under visible light irradiation; (b) XRD patterns of ZnO, $g\text{-C}_3\text{N}_4$, and $g\text{-C}_3\text{N}_4\text{@ZnO}$ samples; (c) UV-vis absorption spectra of ZnO, $g\text{-C}_3\text{N}_4$, and $g\text{-C}_3\text{N}_4\text{@ZnO}$ composites, Copyright © 2022 Elsevier [164]; (d) Malachite Green (MG) photocatalytic activity of the $g\text{-C}_3\text{N}_4\text{/ZnO}$ photocatalyst, Copyright © 2022 Elsevier [165]; (e) the pure ZnO NRAs, $g\text{-C}_3\text{N}_4$ thin film, and $g\text{-C}_3\text{N}_4\text{/ZnO}$ heterostructure photocurrent response to a light on-off under visible light illumination (≥ 420 nm), Copyright © 2022 Elsevier [159]; (f) Nyquist plots of the ZnO, $g\text{-C}_3\text{N}_4$, and $g\text{-C}_3\text{N}_4\text{/ZnO}$ catalysts. Reproduced from Reference [160].

The doping of the heterojunction is a method to change and tune the properties of the bare $g\text{-C}_3\text{N}_4\text{-ZnO}$ heterojunction. The metal elements are one of the excellent candidates used as dopants in the $g\text{-C}_3\text{N}_4\text{-ZnO}$ structure. Ahmad et al. illustrated the effect of different metal dopants on photocatalytic efficiencies [169]. Ag-doped $g\text{-C}_3\text{N}_4\text{-ZnO}$ showed the highest specific area compared to the pure ZnO, $g\text{-C}_3\text{N}_4$, and Al, Mg, Ni, Cu-doped heterojunctions. The redshift is observed for UV-vis absorption for the metal-doped $g\text{-C}_3\text{N}_4\text{-ZnO}$ composite compared to the pure ZnO, revealing the more visible light absorption, and inducing a higher charge carrier generation rate [169]. The bandgap of the ZnO, $g\text{-C}_3\text{N}_4$, Al, Mg, Ni, Cu, and Ag-doped $g\text{-C}_3\text{N}_4\text{-ZnO}$, which is determined by the Tauc's plots, is 3.23 eV, 2.66 eV, 3.15 eV, 3.08 eV, 3.06 eV, 3.0 eV and 3.05 eV, respectively. The PL spectra of these doped composites demonstrated the three emissions at 391 nm, 456–466 nm, and 550 nm, which are attributed to the free exciton transition or recombination process, bandgap recombination of photoinduced charge carriers, and oxygen vacancies [169]. The photocurrent of the metal-doped structure is higher than that of the pure ZnO and $g\text{-C}_3\text{N}_4$. The Cu-doped composite showed the lower electron transition resistance among all composites. It was concluded that the synergistic impact of Cu-doped $g\text{-C}_3\text{N}_4\text{-ZnO}$, promotes electron mobility and separation efficiency [169]. Photocatalytic activity of the Mg-doped composite is higher than that of ZnO, $g\text{-C}_3\text{N}_4$, and $g\text{-C}_3\text{N}_4\text{-ZnO}$ structure. Mg is one of the ideal candidates for charge separation in the composite structure [170]. Other kinds of metal-based materials such as K, Cr, Co, and Fe have shown excellent light absorption, increased photo-induced electron-hole generation and separation, and migration for improved photocatalytic activities [171–174]. ZnO/K@ $g\text{-C}_3\text{N}_4$ had higher stability after five cycles (84%) for tetracycline removal. Doping the Cr into the $g\text{-C}_3\text{N}_4\text{-ZnO}$ structure promoted photocatalytic performance [172]. 60% $g\text{-C}_3\text{N}_4\text{/Cr-ZnO}$ photocatalyst had 93% degradation rate in

1.5 h, which is 3.5, 2.5, and 2-fold higher than that of 5% Cr-ZnO, bulk g-C₃N₄, and 60% g-C₃N₄-ZnO, respectively. Nitrogen is another widely important element used as a dopant for the g-C₃N₄-based heterojunctions [147,175–177]. g-C₃N₄ with 7 wt % of N-ZnO showed the highest photocatalytic RhB degradation with 5 and 4 times higher than those of N-ZnO, and g-C₃N₄, respectively [175]. g-C₃N₄-ZnO with N doping exhibited a 77% higher H₂ evolution rate than that of pure carbon nitride and showed a great charge carrier transfer [147,176]. The N-doped heterojunction revealed the high photocatalytic activities for methylene blue degradation since it shows the narrower bandgap [178]. Other researchers showed the impact of carbon doping in the ZnO-g-C₃N₄ composites [177,179–181]. The Z-scheme heterojunction system containing C-doped g-C₃N₄ grafted on the C, N co-doped ZnO was used to improve the optical properties for enhancing the BPA organic pollutants photodegradation and hydrogen evolution reaction [177]. Oxygen and sulfur are also used to improve the efficiency of the composites [181,182].

Different g-C₃N₄-ZnO heterojunction-based ternary composites have been constructed by various researchers worldwide [183–187]. The surface plasmon resonance (SPR) effect of Ag and Au nanoparticles (NPs), resulting in the improved photocatalytic activities improve by increasing the electron-hole generation and separation, so many researchers tend to use Ag NPs in the heterojunction structures [188–192]. Ag NPs facilitate the migration and improve the photoinduced electron-hole pairs separation by creating close interfaces between g-C₃N₄ and ZnO. Besides, Ag NPs reduce the energy barrier for CO₂ to increase the intermediate radicals on the surface of the nanocomposites [191,193]. It is also revealed that the reaction constant rate for Ag (5 mol%)/ZnO/ g-C₃N₄ is 2.4 times higher than ZnO/ g-C₃N₄. Ag (5 mol%)/ZnO/ g-C₃N₄ composite extends its surface, leading to promoting the photo-generated electron-holes pairs and increasing the lifetime and stability of the charge carrier [194,195]. To show the improved photocatalytic performance with high stability, different Ag-based compounds between g-C₃N₄ and ZnO have been investigated [188,196–199]. In another research, the hydrothermally synthesized Ag-ZnO/S-g-C₃N₄, comprising ZnO NPs doped with 7% Ag with 25% Sulfurized-g-C₃N₄, exhibited outstanding MB photodegradation (97% in 40 min) with excellent recyclability [200]. Carbon-based materials are also used beside the g-C₃N₄-ZnO structure [199,201,202]. Graphene oxide (GO) is carbon-based material commonly used in the g-C₃N₄-ZnO heterojunction [199,203,204]. GO is a two-dimensional platform, providing a promising electron conductivity, high specific surface area, and Young's modulus, which are useful for improving photocatalytic activities. The RhB dye degradation is about 99% for ZnO-g-C₃N₄-GO nanocomposites in 14 min [199]. The ternary nanocomposites provide high stability, which can be used for a wide range of environmental applications [203]. Other forms of carbon-based nanomaterials such as carbon dots (C Dots) are also used to improve the heterojunction structures' efficiency [201,205]. The C Dots provide the facile photoinduced electrons transfer from the ZnO's CB to the g-C₃N₄'s VB. The Z-scheme heterojunction structure can be used in biomedical applications for bacteria-killing and acceleration of wound healing system [201].

5.2.3. Iron Oxide-g-C₃N₄

Fe_xO_y such as FeO, Fe₃O₄, and Fe₂O₃, not only can improve the photocatalytic performance of the g-C₃N₄ structure due to some unique characteristics but they can also be vastly used for contaminant removal in various media [206–209]. Compared to the combination of g-C₃N₄ with TiO₂ or ZnO, fewer papers have been focused on the g-C₃N₄-FeO_x heterojunctions. Xu et al. announced that the CB and VB of Fe₂O₃ are 0.3 eV and 2.4 eV, respectively [210]. FeO_x failed to show any appreciable photocatalytic activity since the improper, more positive CB edge position (Figure 14a). It is confirmed that g-C₃N₄/α-Fe₂O₃ nanocomposites obey a Z-scheme mechanism for photogenerated charge separation. Working on the g-C₃N₄-FeO_x composite was ignited by the pioneering study authored by Ye et al. [211]. They demonstrated that the efficiency of the Fe₂O₃/g-C₃N₄ photocatalysts was increased up to 1.8 times than the bulk C₃N₄ for the RhB degradation under visible light irradiation.

One of the most common synthetic methods to make iron oxides, especially superparamagnetic iron oxides (SPIONs), which show the highest saturation magnetization among all iron oxides while having a low magnetic coercivity, is co-precipitation method [212]. The effect of different parameters such as solution temperature, alkalinity, and stirring rate were investigated by Hosseini's group on the suspension properties for MRI applications [213]. Iron oxides have many applications in different area such as biomedical applications, environmental application, etc. [214,215]. Wang et al. prepared g-C₃N₄-Fe₂O₃ nanocomposite via chemical co-precipitation approach [209]. They also evaluate the adsorption and desorption of seven polycyclic aromatic hydrocarbons (PAHs). Low limits of detection (LOD), excellent linearity, and recovery of the g-C₃N₄/Fe₃O₄ nanocomposites revealing their ideal candidacy for environmental applications, especially PAHs removal from water samples. In another research, the Fe₂O₃-g-C₃N₄ heterojunction was synthesized by thermal treatment in a hypoxia environment [216]. The presence of Fe₂O₃ in the structure will reduce the recombination rate and promote N₂ adsorption. Some other researchers used in-situ thermal condensation to prepared g-C₃N₄ with iron oxides, which can be used in ciprofloxacin (CIP) degradation [217]. CIP is an antibiotic for bacteria sterilization by inhibiting the bacterial DNA. Besides, it is demonstrated that the remnant CIP in the soil can be absorbed by plants and transfer to the human body by consumption. The long-term CIP intake leads to some serious health issues [218]. The hydrothermal method is another method for preparing this composite [219]. In this method, researchers used the solution containing colloidal of the mixture of both Fe₂O₃ and g-C₃N₄. Fe₂O₃ could not cause the methanol yield since the low conduction band of the Fe₂O₃. To be more specific, Fe₂O₃ (5, 10, 15, 20 wt %) was added to 500 mg of g-C₃N₄ (in 25 mL of water), prepared by direct solid-state reaction of dicyandiamide and thiourea, to form a homogeneous mixture. The solution was transferred into the Teflon-lined stainless-steel autoclave at 150 °C for 4 h, and then washed and dried at 60 °C overnight. As a result, Duan and Mei revealed that the g-C₃N₄-Fe₂O₃ heterojunction significantly improved methanol yield from the CO₂ photoreduction.

As mentioned above, Fe₂O₃ has a narrow bandgap of about 2.1 eV, making the iron oxides an excellent candidate for broad visible light absorption. Some factors, such as the high recombination rate of electron-holes, the short diffusion length of holes, and lack of sufficient conductivity, improve the need to constructing a FeO_x-based composite, which will increase the applications of this composite. One of the most well-known FeO_x-based composites is the g-C₃N₄-Fe₂O₃, which can adequately promote photocatalytic activities. Geng et al. illustrated the XRD and FTIR pattern of the g-C₃N₄-Fe₂O₃ composites, which are similar to the results of other publications [220]. Any significant peak shift is observed in the XRD pattern of the composites, which are indicated in Figure 14b. Additionally, the intensity of the peaks related to the (104) and (110) of α-Fe₂O₃ are strengthened by increasing the α-Fe₂O₃ content in the structure [220]. They also investigated that the FTIR characteristic peaks of the g-C₃N₄-Fe₂O₃ composites are not significantly change compared to the bare g-C₃N₄ and Fe₂O₃ (Figure 14c). As mentioned previously, the broad peak around 3500 cm⁻¹ is due to the presence of moisture in the samples. The characteristic peaks at the bare Fe₂O₃ and g-C₃N₄ comprising 3000–3400 cm⁻¹ and unresolved peaks from 1237 to 1640 cm⁻¹ indicating N–H, C–N, and C=N in the g-C₃N₄, respectively [220]. Besides, the sharp peak at 811 cm⁻¹ is related to the stretching vibration of the triazine units. At the same time, the peaks at 543 cm⁻¹ and 469 cm⁻¹ directly correspond to the Fe–O stretching vibrations. The optical studies are also depicted in Figure 14d,e, which not only showed absorption and emission of this composite but also revealed the effect composition on these characteristics features. All samples showed a promising UV absorption above 450 nm compared to the g-C₃N₄, helping the photocatalytic characteristics improvements (Figure 14d) [221]. The PL emission spectra showing in Figure 14e will aid us in a clearer understanding of the photophysical behavior of the prepared materials. The results showed a broad peak at the range of 440–463 nm, while the PL intensity of the Fe_xO_y showed a high decrease. The decrease in the PL intensity may be due to the reduction in the luminous recombination probability, resulting in enhancing the charge

separation and photocatalytic reactions. Cheng et al. also demonstrated the insignificant deactivation in about 16 h during 4 cycles showing high stability of this composition. In order to improve the photocatalytic activities, Wang and his coworkers used Al–O bridged $g\text{-C}_3\text{N}_4\text{-}\alpha\text{-Fe}_2\text{O}_3$ z-scheme nanocomposites [222]. In this work, the photocurrent density with 450 nm excitation wavelength increased when researchers used the Al–O bridged 15 $g\text{-C}_3\text{N}_4\text{-}\alpha\text{-Fe}_2\text{O}_3$, containing 15 mass percentage of $g\text{-C}_3\text{N}_4$ and 6 mole percent of Al to Fe. This analysis indicated in Figure 14f showed that Al–O bridged $g\text{-C}_3\text{N}_4$ facilitated the photo-generated charge transfer and separation [222].

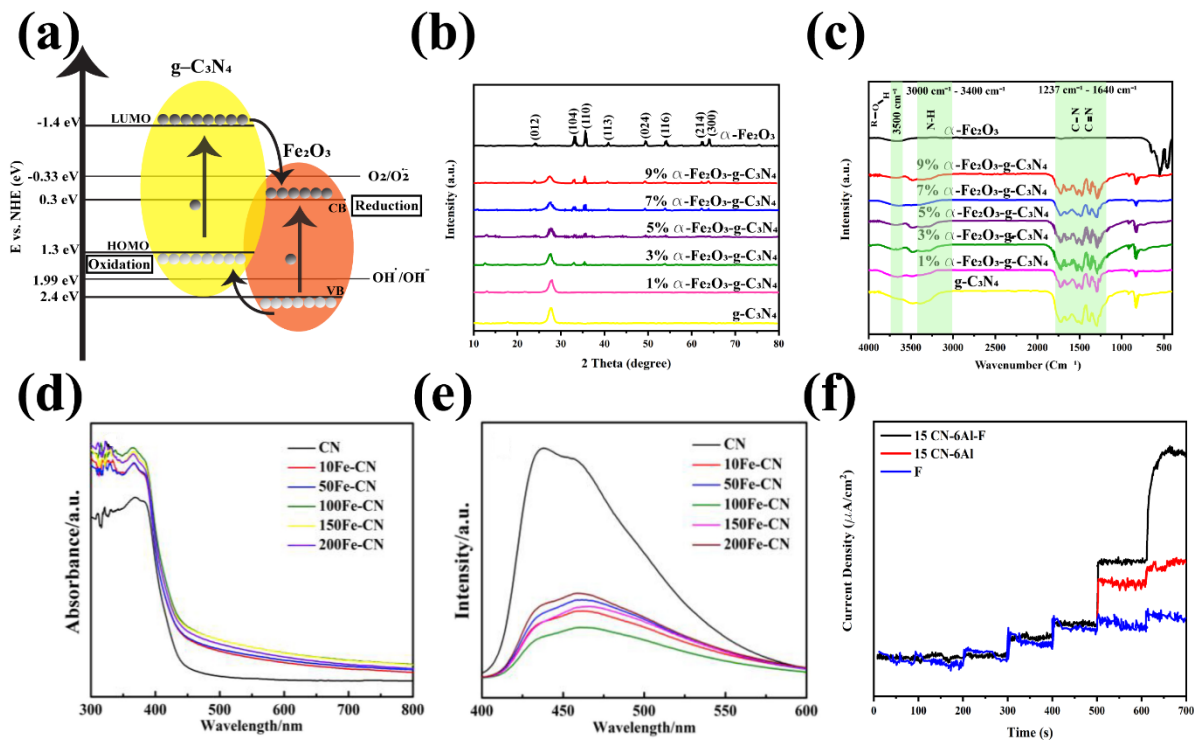


Figure 14. (a) Proposed structure of $g\text{-C}_3\text{N}_4\text{-FeO}_x$ heterojunction interface; (b) XRD patterns and (c) FT-IR spectra of $g\text{-C}_3\text{N}_4$, $\alpha\text{-Fe}_2\text{O}_3$, and $\alpha\text{-Fe}_2\text{O}_3/g\text{-C}_3\text{N}_4$ composites, Copyright © 2022 Elsevier [220]; (d) UV-Vis diffuse reflection absorption spectra; (e) PL spectra of CN and $x\text{Fe-CN}$ samples with an excitation wavelength of 380 nm, Copyright © 2022 Elsevier [221]; (f) single-wavelength photocurrent response of F, 15CN-F and 15CN-6Al-F, Copyright © 2022 Elsevier [222].

Metastable materials that transform from one to another state over a long period of time have superior properties. Bipyrite phase of iron(III) oxide ($\beta\text{-Fe}_2\text{O}_3$) act as its α -phase (hematite) iron(III) oxide. However, it shows a more desirable bandgap (1.8 eV) for photocatalysis. Christoforidis et al. prepared metastable β -phase Fe_2O_3 nanoparticles on the $g\text{-C}_3\text{N}_4$ surface by a solid-state, in-situ growth method, without the need of specialized equipment, surfactants, stabilization, or precipitating agents [223]. In this research, the $\beta\text{-Fe}_2\text{O}_3$ improve the photocatalytic activities by increasing the ability of light absorption in the visible region, and enhanced carriers' separation. The hybrid $\beta\text{-Fe}_2\text{O}_3/g\text{-C}_3\text{N}_4$ nanomaterials are an excellent candidate for photodegradation since they showed higher photocatalytic activity and a promising stability [223].

There are several $g\text{-C}_3\text{N}_4\text{-FeO}_x$ -based composites with promising photocatalytic activities. The $g\text{-C}_3\text{N}_4$ coated with the Fe_xO_y covered by metals will enhance the photocatalytic properties by ameliorating the photo-induced charge generation and separation. The promising LSPR effect of the Au and active sites of Pt convinced researchers to use $\text{Fe}_2\text{O}_3/\text{Pt}/\text{Au}$ nanocomposite immobilized on the $g\text{-C}_3\text{N}_4$ surface as an excellent composite for hydrogen evolution [224]. As discussed in our previous papers, Au nanoparticles with strong LSPR can participate in catalytic reactions (for an example see [225]). Au NPs with a

strong LSPR effect enlarge the photo-electron conversion efficiency of the photocatalyst. The researcher also investigates the CO₂ photoreduction of g-C₃N₄ quantum dots-Au NPs co-modified CeO₂/Fe₃O₄ micro-flowers (MFs). They showed that Au NPs promote photocatalytic activities by generating the electrons and holes and enhance carriers' separation in CeO₂ MFs and CN QDs in the photocatalyst [226]. Copper is another metal used in the g-C₃N₄-FeO_x-based composite. Liu et al. fabricated g-C₃N₄/Fe₂O₃-Cu for electrochemical detection of glucose [227]. g-C₃N₄/Fe₂O₃-Cu composites improved the electrochemical performance for glucose detection with a LOD of 0.3 mM. They also confirmed that g-C₃N₄-Fe_xO_y composite can be used as an electrode in sensors to measure other compounds. The effect of a multi-walled carbon nanotube was investigated by Zhang et al. [228]. They demonstrated that due to the large specific area, hydrogen bonds, π - π , and electrostatic interactions of the MWNTs@g-C₃N₄@Fe₂O₃, these 3D structures were novel magnetic solid-phase extraction sorbents for PAH with the LOD of 0.001–0.5 mgr·L⁻¹. Besides, the 3D structure has a good repeatedly and recovery for 16 PAHs in the water samples. Graphene, as other carbon-based materials also used to improve the efficiency of Fe₃O₄/g-C₃N₄ composites. Wng et al. showed that the Fe₃O₄/graphene/S doped g-C₃N₄ dose of 1.0 g/L comprising 20% Fe₃O₄ mass fraction could completely remove Ranitidine (≤ 2 mg/L) in 60 min, an initial pH of 7.0 [229].

The other ternary composite used for the water splitting is g-C₃N₄/CeO₂/Fe₃O₄ [230]. The composites showed enhanced oxygen and hydrogen evolution reaction with high current density (40 mA·cm⁻²) at the potential of 327 mV, which was greater than the bare Fe₃O₄ and bulk g-C₃N₄. The ternary composites showed excellent stability and negligible activity loss up to 14 h. the Z-scheme g-C₃N₄/Fe₃O₄ can be coupled with the CdS and was used for different antibiotics degradation [231]. The results showed the improved degradation rate to 45 times of CdS, 26 times of pure g-C₃N₄, and 9.5 times of CdS/g-C₃N₄ for the tetracycline removal. The presence of the Fe₃O₄ improves the photocatalytic performance and stability by an increase in the inducing and separation of the electron-hole pairs and generating more ·O₂⁻ for organic pollutant degradation [231].

Other Fe_xO_y compounds are also used to improve the UV-visible light absorption for different applications. NaFe₂O₄ is one of the most well-known FeO_x structures, using in the g-C₃N₄-based composites [232–234]. The Fe₃O₄@NiFe₂O₄-g-C₃N₄ improves photocatalytic activity up to 90% of CIP degradation by reducing the electron-hole recombination [232]. In all superparamagnetic samples, the high M_s assists in improvig of the recyclability and stability of the photocatalyst. The more magnetite content in the sample leads to a higher probability in the particle agglomeration, resulting in the decrease in the active sites of the and the reduction in the photocatalytic efficiencies. The fabricated g-C₃N₄/NiFe₂O₄ can also be used to degrade MB and RhB by activating H₂O₂ to produce the oxidizing reagent [233,234]. The magnetic properties of the NiFe₂O₄ (M_s= 45 emu/g) were induced to the whole composite (M_s= 40 emu/g) to promote repeatability and improve the photodegradation rate [233]. The other most popular compound captured researchers' attentions are ZnFe₂O₄ and LaFeO₃ [235–237].

5.2.4. WO₃-g-C₃N₄

Figure 15a demonstrated the band edge position of the WO₃ compared to the g-C₃N₄. This Figure illustrated that the bandgap of the WO₃ is 2.6 eV with VB and CB of 2.9 eV and 0.3 eV, respectively, which are more positive than that of g-C₃N₄ and the O₂/O₂⁻ potential [238–240]. Thus, it is approximately impossible to produce ·O₂ by using the traditional type-II mechanism. Many researchers showed that the charge migration almost occurred by the Z-scheme mechanism in the binary g-C₃N₄-WO₃ composite [241–243]. Various methods have been fabricated this binary composite. The hydrothermal approach is one of the main routes to prepare the g-C₃N₄-WO₃ composite [244,245]. Zhang et al. prepared WO₃/g-C₃N₄ by dissolving the WCl₆ and ascorbic acid in ethanol and g-C₃N₄ followed by 5 min sonication and stirred for 20 min [244]. The uniform suspension was heat-treated at 220 °C for 12 h, and then washed several times using ethanol. The other

way for the preparation of the heterojunction is the microwave irradiation technique with the direct calcining of the WO_3 and $\text{g-C}_3\text{N}_4$ combination at $400\text{ }^\circ\text{C}$ for 2 h [246]. In this method, to prepare WO_3 by simple household microwave irradiation, tungstic acid in NaOH solution was mixed and stirred for 30 min to form a tungstic hydroxyl group [246]. Afterward, the pH of the prepared mixture was reduced to 1 by adding HCl solution, and the gel should be put into the Teflon-lined household microwave oven (2.45 GHz) for 10 min to prepared WO_3 . Finally, the mixture should be placed in an alumina crucible with a cover in a muffle furnace and heated at $400\text{ }^\circ\text{C}$ for 4 h. A wet chemical process, sonochemical, in situ self-assembly, etc., are other ways to synthesize the $\text{g-C}_3\text{N}_4$ - WO_3 heterojunction [247–249].

$\text{g-C}_3\text{N}_4$ - WO_3 composites were characterized by various techniques. In most research papers, XRD and FTIR are the prime characteristic methods to evaluate the formation of $\text{g-C}_3\text{N}_4$ - WO_3 . Researchers announced that the WO_3/CN wt % = 10% provides the best characteristics compare to the bulk $\text{g-C}_3\text{N}_4$ and WO_3 [250]. Figure 15b shows the XRD patterns of the bulk $\text{g-C}_3\text{N}_4$, WO_3 , and $\text{g-C}_3\text{N}_4$ - WO_3 , perfectly showing the impact of WO_3 content on this characterization. There are nine distinct peaks for the as-prepared WO_3 [247,251]. Additionally, the binary composites revealed the combination peaks of the WO_3 and $\text{g-C}_3\text{N}_4$. The higher WO_3 content leads to the lower peak's intensity of the $\text{g-C}_3\text{N}_4$, corresponding to the expansion of the interlayers and $\text{g-C}_3\text{N}_4$ coverage with a WO_3 . The low intensity of the peaks at 23.5° and 36.6° , which is detectible in the sample with a high WO_3 to $\text{g-C}_3\text{N}_4$ ratio, is related to the hexagonal-phase WO_3 and illustrated the formation of the composite successfully. They also characterized the $\text{g-C}_3\text{N}_4$ - WO_3 composite with the FTIR spectra analysis [251]. Like other materials' FTIR spectra, the wide peak at the range of $3000\text{--}3500\text{ cm}^{-1}$ in all samples is attributed to the N-H and O-H stretching vibration, and the adsorbed water molecules' bending vibration stands at about 1630 cm^{-1} for samples (Figure 15c). The broad peak at the $750\text{--}1000\text{ cm}^{-1}$ corresponds to the O-W-O stretching vibrations of WO_3 [252,253]. As a result, although the $\text{g-C}_3\text{N}_4$ characteristic FTIR peaks can be observed in the composite FTIR spectra, WO_3 peaks are not significantly detected in the hybrid composite, which can be ascribed to the vacancies between the $\text{g-C}_3\text{N}_4$ clusters or band overlapping. Optical studies were carried out by Chai et al. on the $\text{g-C}_3\text{N}_4$ with WO_3 structure [254]. The UV-vis DRS of the WO_3 , $\text{g-C}_3\text{N}_4$, and WO_3 - $\text{g-C}_3\text{N}_4$ composites with the WO_3 contents is depicted in Figure 15d. The obvious absorption edges at $\sim 470\text{ nm}$ and 455 nm are detected for WO_3 , $\text{g-C}_3\text{N}_4$, respectively, corresponding to the 2.64 eV and 2.73 eV. The $\text{g-C}_3\text{N}_4$ - WO_3 composites present the combination absorption features of $\text{g-C}_3\text{N}_4$ and WO_3 [254]. Figure 15e displays the PL emission peak of the bulk $\text{g-C}_3\text{N}_4$ and 18.6 wt % $\text{WO}_3/\text{g-C}_3\text{N}_4$ hybrid composite [254]. The result demonstrates that the intensity of the composites' PL peak at $\sim 440\text{ nm}$ is lower than that of the pure $\text{g-C}_3\text{N}_4$ [255–257]. The PL spectra revealed that WO_3 apparently suppresses the photoinduced electron-hole recombination in the $\text{WO}_3/\text{g-C}_3\text{N}_4$ composites and confirms the Z-scheme interface contact [258]. They are also revealed that the OH^- generating during the photocatalytic reaction leads to the higher PL intensity increases the irradiation time. Besides, the transient PL decay trace of the bare $\text{g-C}_3\text{N}_4$, WO_3 , and $\text{g-C}_3\text{N}_4$ - WO_3 composite hollow microsphere are $\sim 4.46\text{ ns}$, 1.62 ns , 2.23 ns , respectively [259]. The zeta potential values of the WO_3 , $\text{g-C}_3\text{N}_4$ and their binary composites announce the negatively charged surface of the samples [260]. The presence of WO_3 in the composite changes the zeta potential from -5.7 mV to -33.1 mV , while it decreases the BET specific area from $100.97\text{ m}^2\cdot\text{g}^{-1}$ to $47.88\text{ m}^2\cdot\text{g}^{-1}$, which are high enough for promising adsorption ability and photocatalytic activities. $\text{g-C}_3\text{N}_4$ - WO_3 reveals high stability and repeatability after 4 cycles with a slight efficiency decrease for the degradation of RhB [261–263].

There are also some dopants and compounds used to improve the photocatalytic efficiency of the binary composites. In the z-scheme C or Pt- $\text{g-C}_3\text{N}_4$ with hydrogen treated WO_3 , the electrons from $\text{g-C}_3\text{N}_4$ and holes from WO_3 facilitate the photogenerated charge carries generation, which will enhance the photocatalytic activities [254,264]. In other words, the C or Pt dopants help the composite to improve the light absorption and charge

separation. This structure also enhances stability and repeatability. O-doped $g\text{-C}_3\text{N}_4\text{-WO}_3$ has 4.7 times higher H_2 performance than the bare composite [265]. The carbon vacancies and $g\text{-C}_3\text{N}_4$ oxidization resulted in the formation of the porous composites and a decrease in the composite's surface area. The effective Si-O bridge between $g\text{-C}_3\text{N}_4$ and WO_3 significantly promotes charge transfer and separation [266].

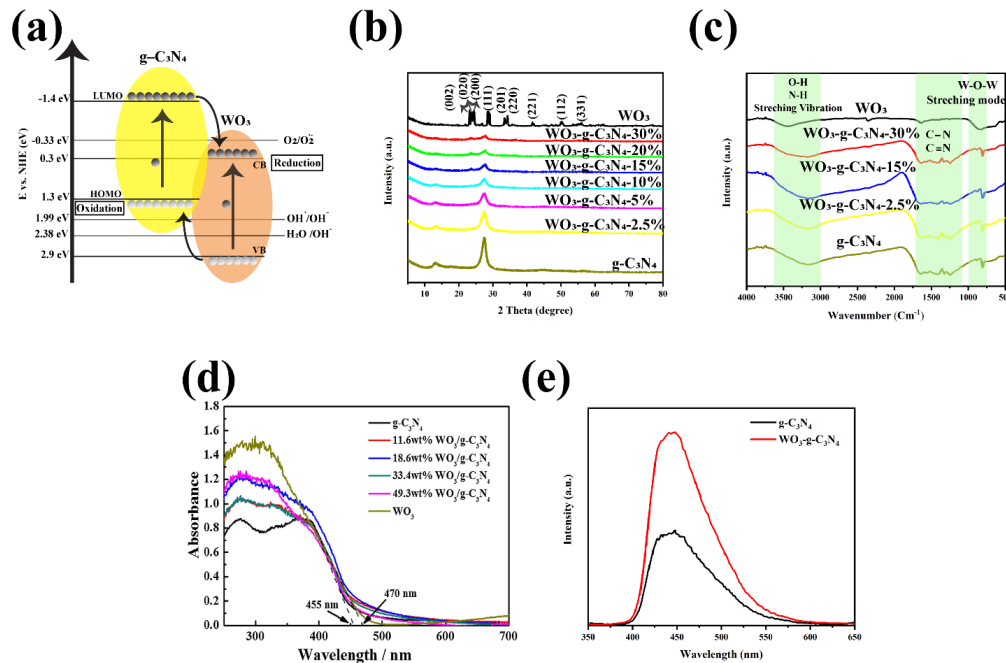


Figure 15. (a) Proposed structure of $g\text{-C}_3\text{N}_4\text{-WO}_3$ heterojunction interface; (b) XRD patterns and (c) FT-IR spectra of obtained samples (x in $\text{WO}_3/g\text{-C}_3\text{N}_4$ refer to the mass ratio of WO_3 to $g\text{-C}_3\text{N}_4$), Copyright © 2022 Elsevier [251]; (d) UV-DRS spectra of pure $g\text{-C}_3\text{N}_4$, WO_3 and $\text{WO}_3/g\text{-C}_3\text{N}_4$ heterojunction; (e) PL emission spectra of $g\text{-C}_3\text{N}_4$ and 18.6 wt % $\text{WO}_3/g\text{-C}_3\text{N}_4$ composite, Copyright © 2022 Elsevier [254].

In addition to the doped binary composites, adding other compounds to form the ternary structure is a practical way to boost the composite characteristics. Mediators and co-catalyst such as metals (such as Ag, Au, Cu, Pt, and Sn) are currently used to improve the Z-scheme composites by facilitating the transporting and carrier capturing during the photocatalyst process [267–272]. Li et al. demonstrated the charge carrier migration between various $\text{WO}_3\text{-Metal}$ (Cu, Ag, Au)- $g\text{-C}_3\text{N}_4$. They showed that Cu plays the ideal candidate for photocurrent enhancement and improves the photocatalytic performance in the Z-scheme $g\text{-C}_3\text{N}_4\text{-WO}_3$ heterojunction among different metals used in the research. In other words, the Cu- $g\text{-C}_3\text{N}_4$ and $\text{WO}_3\text{-Cu}$ are favorable for electron migration since they have the matched Fermi level of energy [267]. In other research, the developed $\text{WO}_3/\text{Ag}/g\text{-C}_3\text{N}_4$ ternary composite was used for the RhB and tetracycline (TC) degradation. The results demonstrated that the improved photocatalytic activity of $\text{WO}_3/\text{Ag}/g\text{-C}_3\text{N}_4$ is obtained due to the large contact region between $g\text{-C}_3\text{N}_4$ nanosheets and WO_3 nanoplates. Besides, the presence of the Ag NPs in the composite with SPR effect accelerates the charges to transfer, improves the photocatalytic activity, and enhances the stability and repeatability [268,269].

Researchers also worked on the $\text{Ag}/g\text{-C}_3\text{N}_4/\text{WO}_3$ to degrade oxytetracycline hydrochloride under visible light [273]. The 0.4 g/L of $\text{Ag}/g\text{-C}_3\text{N}_4/\text{WO}_3$ composite demonstrated the highest photocatalytic activity, which could degrade 97.74% of oxytetracycline (10 mg/L) in 60 min. In another research, Qin et al. showed that the presence of Pt in the $\text{WO}_3\text{-}g\text{-C}_3\text{N}_4$ composites possesses excellent photocatalytic H_2 evolution with 1299.4 μmol under visible light, which is higher than that of $\text{WO}_3/g\text{-C}_3\text{N}_4/\text{Pt}$ and pure CN with

1119.4 μmol , and 113.2 μmol , respectively [274]. As mentioned above, $\text{g-C}_3\text{N}_4/\text{Au}/\text{WO}_3$ as Z-scheme heterojunction displayed excellent photocurrent that can be used in photoelectrochemical immunosensing of Aflatoxin B1 in food, which can be dangerous for humans [275]. The methane formation over the $\text{WO}_3\text{-Bt-g-C}_3\text{N}_4$ composite was 5.98, 6.74, and 25.19 times higher than that of $\text{WO}_3\text{-g-C}_3\text{N}_4$, $\text{Bt-g-C}_3\text{N}_4$, and $\text{g-C}_3\text{N}_4$ samples, respectively [272].

Some promising properties of tungsten-based materials such as electrical, optical, magnetic, and photocatalytic activities, low-cost preparation persuade researchers to use these materials such as NiWO_4 , BaWO_4 , CuWO_4 , and BiWO_6 [270,276–278]. The band structures of these compounds with $\text{g-C}_3\text{N}_4$ and WO_3 make the CB and VB positions match each other, leading to the prolonged charge carriers' lifetime in the form of a double Z-scheme system. The photoelectrochemical (PEC) of a double Z-scheme $\text{g-C}_3\text{N}_4\text{-WO}_3\text{-Bi}_2\text{WO}_6$ system reveals the enhanced photocurrent density, reduced electron-hole recombination, resulting in the promotion of photocatalytic efficiency [276]. Other materials have been anchored in the $\text{g-C}_3\text{N}_4\text{-WO}_3$ to form an excellent composite for different applications [279,280]. Bi-based materials are among the prime and important materials used to construct the composites for photocatalytic activities [281–283]. MoS_2 and MoO_3 are other compounds to be utilized as materials for this ternary composite [284,285]. $\text{g-C}_3\text{N}_4/\text{WO}_3$ with TiO_2 can also be useful for the methylene blue dye degradation, which its efficiency is about 3.1 folds higher than that of the binary counterparts of TiO_2/WO_3 (0.00691 min^{-1}) in 120 min. The kinetic constant for different composition of this composite are in order of TWG-15\% (which 15% is the $\text{g-C}_3\text{N}_4$ wt %) > TWG-10\% > TWG-5\% > TWG-20\% > TW [286].

5.2.5. Tin Oxide- $\text{g-C}_3\text{N}_4$

Other metal oxides widely used in $\text{g-C}_3\text{N}_4$ binary heterojunction are tin oxides. SnO (stannous oxide) and SnO_2 (stannic oxide) are two forms of tin oxide. Figure 16a shows the band edges' position in the tin oxide and compares it to the CB and VB of the $\text{g-C}_3\text{N}_4$. He et al. showed that the SnO_2 has a large bandgap of about 3.6 eV, and its CB level is lower than that of $\text{g-C}_3\text{N}_4$ [287]. It is also demonstrated that these tin oxide- $\text{g-C}_3\text{N}_4$ composites mostly form the Z-scheme heterojunction [288,289]. Tin oxide can be designed with various shapes by wet and dry methods. The heterojunction is synthesized by different synthetic methods, including hydrothermal and thermal treatment [78,290–292]. Sadrnezhad and coworkers prepared $\text{g-C}_3\text{N}_4\text{-SnO}_2$ with the pyrolysis of urea under microwave irradiation [293]. In this work, tin, ammonium, and urea were put into the beaker and placed in a microwave oven operating at 2.45 GHz and 900 W for 30 min. Finally, the product was washed and dried in the oven. In another work, the mesoporous SnO_2 decorated with $\text{g-C}_3\text{N}_4$ was prepared via pulsed electrophoresis and facile water-crystallization [293]. With pulsing electrophoresis optimized parameter, the 0D $\text{g-C}_3\text{N}_4$ can be homogeneously and completely distributed inside the 1D SnO_2 . Sol-gel is also used to fabricate an efficient $\text{g-C}_3\text{N}_4\text{-SnO}_2$ photocatalyst [294].

Several techniques have been used to confirm the formation of this composite. The XRD pattern and FTIR spectra of $\text{g-C}_3\text{N}_4$, SnO_2 , and $\text{SnO}_2\text{-g-C}_3\text{N}_4$, are depicted in Figure 16b,c [295]. SnO_2 does not change during the preparation of the final structure, which can be seen from the insignificant differences between the pure SnO_2 and the $\text{SnO}_2\text{-g-C}_3\text{N}_4$ XRD pattern in Figure 16b. The characteristic peaks in SnO_2 revealed the (110), (101), (200), and (211) planes of a tetragonal rutile-like structure. Other small peaks, which are not mentioned in Figure 16b, are related to the (220), (310), (301), (202), and (321). As shown in Figure 16b, the (002) crystal plane peak of $\text{g-C}_3\text{N}_4$ overlapped with the (110) crystal plane peak of SnO_2 . Additionally, the Sn–O–Sn anti-symmetric stretching vibration between 400 cm^{-1} and 700 cm^{-1} was demonstrated in Reference [295] (Figure 16c). The red-shift peak at 597 cm^{-1} was a good indication of the formation of $\text{SnO}_2\text{-g-C}_3\text{N}_4$ heterojunction [296,297]. The presence of SnO_2 in the structure suppresses the electron-hole recombination rate. Peng et al. demonstrated the negligible differences and slight blue shift of $\text{g-C}_3\text{N}_4\text{-muscovite sheet}/\text{SnO}_2$ structure, compared to the absorption edges of $\text{g-C}_3\text{N}_4$ (Figure 16d) [298]. Introducing the muscovite sheets does not affect the composites'

absorption, and g-C₃N₄-muscovite sheet/SnO₂ absorption spectra reveal the potential photocatalytic applications under visible light. They also showed that the g-C₃N₄-muscovite sheet/SnO₂ Cement has an excellent photocatalytic activity for RhB stains and isopropyl alcohol photodegradation. Due to the low electron-hole recombination probability of the SnO₂-g-C₃N₄, the intensive PL intensity was observed in Figure 16e [298,299]. The faster interfacial charge-transfer and lower resistance, results in the Nyquist plot diameter decrease compared to SnO₂ and g-C₃N₄, shown in Figure 16f [300]. This is therefore a promising photocatalyst. The properties and morphology of this composite change when the relative content of g-C₃N₄ varies in the heterojunction [301,302]. Chen et al. showed that the g-C₃N₄ mass ratio of 72% provides the highest photocatalytic performance, which is 17 times higher than bulk g-C₃N₄ [303]. SnO₂-g-C₃N₄ heterojunction reveals notable stability and recyclability [18]. In addition to robust photocatalytic activities, this composite has a noticeable photoelectrochemical performance under visible light. Doping different elements, such as Sb, S, B, and P, is one of the main approaches to improve the performance of the SnO₂-g-C₃N₄, which can modify the band edges [304–306].

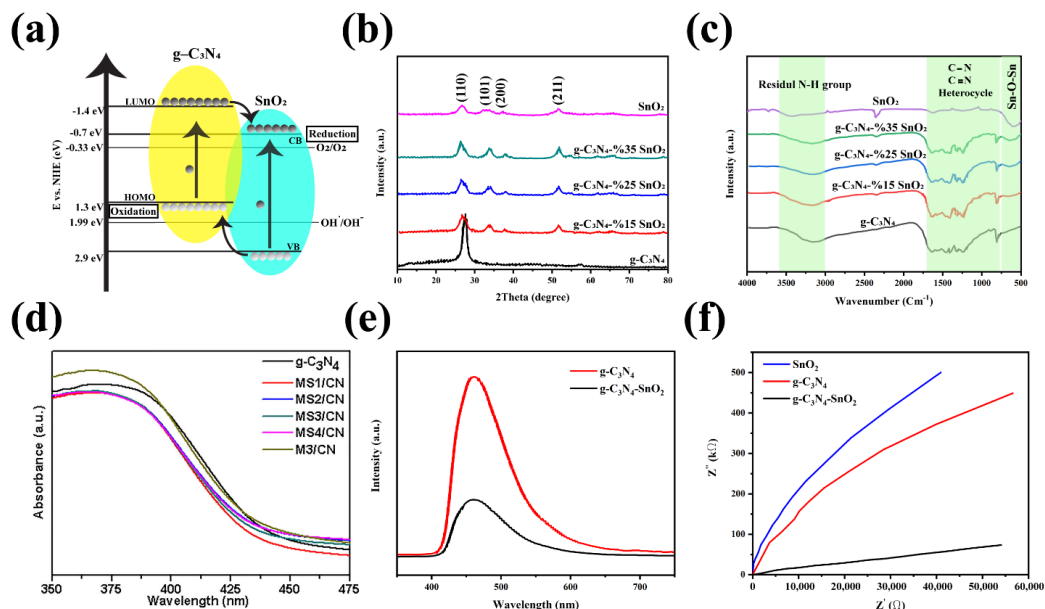


Figure 16. (a) Proposed structure of g-C₃N₄-SnO₂ heterojunction interface; (b) XRD patterns and (c) FT-IR spectra of SnO₂, graphene-like C₃N₄, SnO₂/g-C₃N₄ composites (reproduce from Reference [295]); (d) UV-vis absorption spectra of g-C₃N₄-based samples (muscovite sheet(x)/SnO₂/g-C₃N₄ (MS_x/CN), which x refers to the mass of MS powder), Copyright © 2022 Elsevier [298]; (e) PL spectra of g-C₃N₄ and SnO₂ quantum dots-g-C₃N₄ nanocomposite, Copyright © 2022 Elsevier [299]; (f) EIS Nyquist plots of the bare SnO₂, g-C₃N₄, and SnO₂-g-C₃N₄ structure, Copyright © 2022 Elsevier [300].

Doping is a method to improve the efficiency of SnO_x-g-C₃N₄ heterojunctions, which are used in different applications. SnO₂/g-C₃N₄ sensor doped with Ni shows LOD about 1.38 ppb and has high stability, promising selectivity, rapid response and recovery time, and great resistivity against humidity [307]. Besides, researchers used made Ce doped SnO₂/g-C₃N₄ to make capacitors with a specific capacity of <274 F/g. A supercapacitor with energy and power densities of 39.3 W h kg⁻¹ and 7425 W kg⁻¹, respectively, was made by using Ce-SnO₂/g-C₃N₄/Activated Carbon. The supercapacitor device exhibited retention of 84.2% after completing 5000 cycles [308].

Additionally, several researchers used the SnO₂-g-C₃N₄ based ternary composites to increase the applications of these kinds of composites [309,310]. The plasmonic Au-SnO₂-g-C₃N₄ photocatalyst was fabricated for H₂ evolution and degradation of the organic pollutant, which is commonly used due to the outstanding photocatalytic activities and

significant stability (remain unchanged after 5 h in 5 cycles) [309,311]. In this ternary heterojunction, the presence of the plasmonic Au and g-C₃N₄ offer enhanced photogenerated electrons to the structure. SiO₂ is another compound used besides the SnO₂-g-C₃N₄ system for pollution treatment applications [312]. As mentioned above, TiO₂ has promising properties that aid us in promoting the properties of this composite [313,314]. This ternary composite also demonstrated the perfect antibacterial activity for the degradation of *E. coli* bacteria, probably due to the interface between g-C₃N₄-SnO₂ and TiO₂ and lower charge recombination rate [314]. SnO₂/chitosan/g-C₃N₄ nanocomposite has been used as an Electrochemiluminescence aptasensor to improve lincomycin detection [315]. Ali et al. also showed the cost-effective prepared g-C₃N₄/rGO/SnO₂ nanocomposite for RhB degradation. The optimal amount of this nanocomposite reveals an increased RhB degradation efficiency [316].

As a result, the SnO₂-g-C₃N₄ structure can be used in a wide range of applications. The exploitation of new clean energy instead of fossil fuels is of great interest to many since the fossil energy is exhausted. As a case in point, water splitting is a perfect example of this clean energy generation, which is widely applied by the g-C₃N₄-SnO₂ [317,318]. Besides, in order to reduce environmental pollution, the demand for electric transports has been increased. Thus, it is essential to use the rechargeable batteries such as lithium-ion batteries (LIBs) and improve lithium storage capacity. Specifically, g-C₃N₄ enables SnO₂ anode to enhance the Li storage in these batteries [319,320]. This heterojunction is also used to detect different compounds. Cao et al. showed that SnO₂/g-C₃N₄ composite promoted the sensitivity and selectivity in ethanol gas-sensing applications [321]. The degradation of the inorganic pollutant is another application of this heterojunction [322,323]. The emission of nitrogen dioxide (NO₂) and nitric oxide (NO) causing some environmental issues is one of the biggest challenges among several researchers, solved by Zou and coworkers by using SnO₂-g-C₃N₄ photocatalysts using visible-light irradiation under 30 min [287]. SnO₂-g-C₃N₄ is also used to decomposed Ammonium Perchlorate (AP), a toxic inorganic material [324].

5.2.6. Other Metal Oxides

Other kinds of metal oxides such as V₂O₅, NiO, MoO₃, Cu₂O, Co₃O₄, CeO₂, Bi₂O₃, Al₂O₃, etc., are also used to improve the performance of the bulk g-C₃N₄ [325–345]. The g-C₃N₄-based heterojunctions can be modified by combining with several metals or doping with various agents [346–348]. Cu is a conventional metal using for the improvement of photocatalytic performance. Zhou et al. used Cu/Al₂O₃/g-C₃N₄ for Rhodamine B degradation by H₂O₂ [349]. The Cu immobilized Al₂O₃/g-C₃N₄ also showed promising stability for the treatment of water pollution. Besides, copper is used as a charge separation center for hydrogen evolution, MO, and phenol solution degradation under visible light [350,351]. In addition to Cu, the noble metal Ag and Au is another metal catalyst used besides the g-C₃N₄-based composite [347,352–356]. The Ag and Au can prevent rapid recombination probability, improve the transfer of a generated electron, and enhance the visible light absorption by the surface plasmon resonance, and can be used for a wide range of applications, especially decontamination of organic pollutants. Bi, Pd, Pt, Ni, and Cd are other metal photocatalysts used for improved photocatalytic activities [357–363]. Some semiconductors are also used besides the metal oxide-g-C₃N₄ based composites. Carbon-based nanomaterials provide excellent stability, cost-effective synthesis, enhanced photogenerated electron reservoirs used in bioimaging and sensing, photocatalysis, electrocatalysis [214,336,364–367]. Xie et al. fabricated the carbon quantum dots modified with MoO₃-g-C₃N₄ and demonstrated that this structure showed outstanding visible-light absorption used to degrade tetracycline (TC) from the environment [364]. Besides, among all carbon-based nanomaterials, reduced graphene oxide (rGO) has captured intensive attention [368,369]. Gong and coworkers illustrated that the charges transfer between g-C₃N₄ and Bi₂Fe₄O₉ (BFO) was improved by using rGO. The presence of the rGO causes the separation of electrons and holes in the CB of g-C₃N₄ and the VB of BFO, respectively [354].

In addition to the mentioned metals and semiconductors, metal-organic frameworks (MOFs) are a novel class of porous and crystalline materials with a large surface area-to-volume ratio, high porosity, and tunable pore size that might improve the biosensor sensitivity. Since these structures are made from metal ions, clusters, and organic ligands, these materials can promote the separation and transfer of photoinduced electrons, making them a promising candidate for photocatalytic activities, such as organic pollutants degradation, water splitting, and CO₂ reduction [370]. Cui et al. utilized the Fe-based MOFs, MIL-53(Fe), with Bi₂O₃ and g-C₃N₄ for the degradation of amino black 10B since this structure can enhance the visible light absorption range [371]. Like other heterojunctions, doping is another element to improve the performance of these composites and widen their applications [372–376].

Some g-C₃N₄-based ternary composites comprising the metal oxide compounds can promote photocatalytic activities [103,377–380]. Bismuth complex oxides are among the most efficient catalysts with layered structures beside the g-C₃N₄-based composites. Among various Bi-based compounds, Bi₂O₃ based catalysts have drawn significant attention in the different areas [381,382]. It illustrated that CuO₂/Bi₂O₃/g-C₃N₄ nanocomposite reveals improved photocatalytic activities for decomposing of 2,4-dichlorophenol under visible light [381]. In another research work, Vattikuti et al. prepared Bi₂O₃/V₂O₅ photocatalysts anchored on the g-C₃N₄ nanostructure, which can be used for the phenol red (PR) pollutant degradation [383]. They also demonstrated that the efficiency of hybrid composites for the PR removal under the simulation solar light irradiation was higher than that of fabricated materials. Bi₂O₃/g-C₃N₄ heterojunctions were also conjugated with the BiPO₄ [15]. g-C₃N₄/Bi₂O₃/BiPO₄ hybrid exhibited the perfect photoelectric performance, which is mainly due to the high separation and photogeneration charges and can increment the oxidation/reduction rate. Bi₂O₂CO₃ is another Bi contained material that shows promising photocatalytic activities [384,385]. Kumar and coworkers suggested novel magnetic g-C₃N₄/Bi₂O₂CO₃/CoFe₂O₄ heterojunction with high visible light absorption for reduction of 4-nitrophenol into 4-aminophenol [386]. TiO₂-g-C₃N₄ based ternary composites are widely used for photodegradation. Min et al. fabricated the Cu₂O-TiO₂/g-C₃N₄ hybrid composite for the organic dyes' discolorations. Besides, this nanocomposite can also illustrate good performance for discolorations of RhB, MB, and MO within 3, 10, and 15 min, respectively [387]. TiO₂-g-C₃N₄ composites are also anchored with CeO₂ and metal to form g-C₃N₄-Meⁿ⁺¹/CeO₂-TiO₂ for photooxidation of toluene [388].

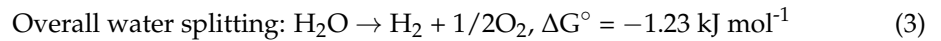
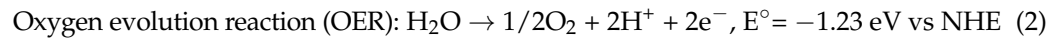
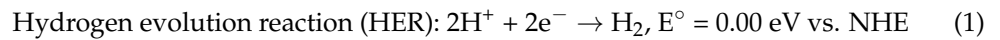
6. Application of Metal Oxide-Based g-C₃N₄ Nanocomposites

6.1. Photocatalysts

6.1.1. H₂ Generation via Water Splitting

These days, the demand for safe, efficient, and renewable energy resources instead of limited fossil fuel sources has increased among more and more people [14,184,389–391]. This replacement is an efficient remedy for global warming and greenhouse gases emission. The hydrogen energy content is in the range of 120 to 142 MJ kg⁻¹, which is higher than that of hydrocarbon fuels. Thus, it is estimated that hydrogen will be responsible for 90% of energy production by 2080. As a result, H₂ generation is a novel and environmentally-friendly research topic among many researchers [8,13,392–395]. One of the most recent hydrogen production techniques is the photocatalytic water splitting method via metal oxide-g-C₃N₄ heterojunctions by using prolific light sources [365,390,396–400]. For water splitting, the band position of the photocatalysts should be modified to provide the CB position more negative than the H₂O reduction potential (0 eV vs. Normal Hydrogen Electrode (NHE)) for H₂ generation and more positive than the H₂O oxidation potential (1.23 eV vs. NHE) for O₂ generation. To be more specific, the generated electrons are used for the hydrogen evolution reaction (HER) and oxygen evolution reaction (OER)

via Equations (1) and (2), respectively, and the final water-splitting reaction is shown in Equation (3) [111].



where NHE is the normal hydrogen electrode.

Generally, there are three steps for each photocatalytic reaction; initially, the semiconductor absorbs light with energy equal to or higher than the bandgap to generate the electrons and holes in the valence and conductive band, respectively. Then, the photoinduced electrons and holes are moved to the surface of the semiconductor to start the reaction. Finally, the charge carriers participate in the reduction and oxidation reactions on the surface of the photocatalysts.

Considering the band edges position of some of the metal oxide-g-C₃N₄ composites, which were noted previously, some heterojunctions are more anodic than that of H₂O reduction potential to show excellent performance under visible light irradiation [111,401]. As a case in point, TiO₂-g-C₃N₄ heterojunctions are widely used as an excellent photocatalytic for H₂ evolution. Yan et al. demonstrated that the efficiency of the visible-light-induced H₂ evolution of the binary composite comprising anatase TiO₂ and g-C₃N₄ was enhanced, and this is due to the desirable photoinduced carriers' separation [402]. In order to raise the heterojunction efficiency, other kinds of materials with various effective characteristics are also used. These materials (metals) or cocatalysts such as Ag, Au, Pt, etc., can host active sites for H⁺ reduction [403,404]. The loading Au and Ag would be significantly beneficial for this application because of their plasmonic characteristic. Marchal et al. illustrated that the optimized components ratios and contact quality in Au/(TiO₂-g-C₃N₄) lead to the enhanced visible light absorption with the proper band positions for photogenerated charge carriers [403]. Besides, the presence of Au and Ag will promote the water splitting for H₂ production by the improved charge separation rate. The H₂ generation rate under sunlight irradiation as a function of the relative ratio of methanol as a sacrificial agent and TiO₂/g-C₃N₄ is depicted in Figure 17a. It was mentioned that no H₂ generation was observed for the Au-free g-C₃N₄ and TiO₂. Furthermore, the best photocatalytic H₂ production was observed for the 0.5 wt % Au/(TiO₂-g-C₃N₄) (95/5) structure, using 1 vol% of CH₃OH as a sacrificial agent (Figure 17a). Carbon quantum dots (CQDs) can also improve photocatalytic performance to effectively decompose H₂O₂ to H₂O and O₂. CQDs possess up-conversion fluorescence spectra, which could convert visible light to ultraviolet or near-ultraviolet light, resulting in excellent photocatalysis. Consequently, it is proven that the combination of C₃N₄, TiO₂, CQDs is a great candidate for water splitting [405]. Other metal oxide-g-C₃N₄ based composites such as WO₃-g-C₃N₄ and ZnO-g-C₃N₄ are used for H₂ generation. Mahala et al. demonstrated that the prepared ZnO nanosheets decorated with g-C₃N₄ quantum dots composites on the fluorine-doped tin oxide (FTO) coated glass slide could be utilized as a photoanode for water splitting via PEC (Figure 17b) [406]. In another work, the effect of boron addition and carbon nitride content did increase the H₂ evolution up to 85% compared to the bare TiO₂, which is mainly due to charge carriers' generation and separation [109]. It was also shown that the photoconversion efficiency of the low charge-transfer resistance of ZnO decorated with g-C₃N₄ is 2.3 times higher than that of pure ZnO [406]. As discussed previously, this composite had a high specific surface area, promising electronic conductivity, and excellent charge transfer interfaces and would be excellent candidates for the water splitting and H₂ evolution [407]. Other materials with suitable properties can also be used to improve the H₂ generation of g-C₃N₄-metal oxide-based composites [141,283].

In a plasmonic photocatalyst, a metal nanostructure with a size less than the wavelengths of the light, is embedded onto dielectric or semiconductor materials [408]. In such systems, the light can lead to a local electromagnetic field by generating localized surface

plasmon resonance (LSPR) and hot carriers. The hot carriers move to conductive and valance bands and this phenomenon is called the LSPR sensitization [409,410]. The hot carriers can be useful for direct oxidation or reduction of chemical species. Some of the metals that showed plasmonic effects are gold (Au), silver (Ag), copper (Co), and platinum. Zhao et al. state that the LSPR of Au can enhance the light absorption and increase the number of photogenerated carriers in the Au/g-C₃N₄/CeO₂ plasmonic heterojunction. The heterojunction was employed for Cr⁶⁺ reduction and oxytetracycline hydrochloride (OTH) catalytic degradation [411].

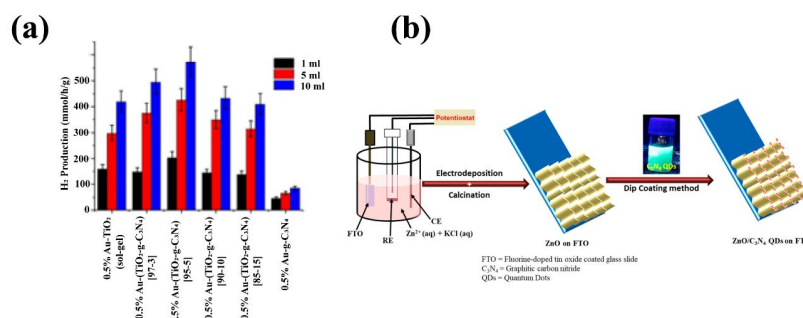


Figure 17. (a) Hydrogen production rate of different samples as a function of the amount of methanol added as a sacrificial agent under solar light irradiation, Reproduced from Reference [403]; (b) schematic image of the ZnO/C₃N₄ QDs synthesis on FTO using electrodeposition and a dip-coating method, Copyright © 2022 Elsevier [406].

Metal oxide-g-C₃N₄ based ternary composites capture lots of attention among researchers [412]. For example, g-C₃N₄ (CN)/TiO₂ (TO)/PbTiO₃ (PTO) films were prepared by the sol-gel followed by the CVD method and were investigated for PEC water splitting [413]. Wang et al. showed the decreased resistance of the interface (R_{CT}) of CN0.10/TO0.4/PTO compared to the pristine PTO and CN0.10/PTO, which facilitated the charge transfer and reduced electron-hole recombination [413]. The incident photo-to-current conversion efficiency (IPCE) was calculated to measure the PEC performance. The pristine PTO has a lower IPCE than CN0.10/PTO, which is mainly due to the higher charge recombination rate. After the TO buffer layer's insertion, the IPCE value was drastically increased to 14.2% at 380 nm [413]. The inserted compact TiO₂ buffer layer provided the type II and Z-scheme interfaces between PTO, TO, and CN and promoted the PEC performance with an improved current density of $-68.5 \mu\text{A cm}^{-2}$ at 0 V versus Ag/Ag-Cl electrode. The high performance is mainly due to the high photogenerated ability of g-C₃N₄/TiO₂ heterojunction. BiVO₄ is another cost-effective compound with a proper bandgap (2.4 eV) to enhance the PEC water splitting [184]. Like PbTiO₃, BiVO₄ can also improve the photocurrent density of the g-C₃N₄@ZnO/BiVO₄ heterojunction to the 0.65 mA cm^{-2} at 1.23 V versus Ag/Ag-Cl electrode.

Table 2 provides some other research activities on the water-splitting application of the g-C₃N₄-metal oxide-based composites.

6.1.2. CO₂ Reduction

CO₂ emission is one of the leading environmental problems causing by fossil fuel consumption and results in a temperature rise of the earth's surface. Photocatalytic CO₂ reduction is a green method to deal with this problem for two reasons [111]. Not only CO₂ reduction reduces the CO₂ emission, but it also solves the future energy demands by producing energy fuels such as CH₄, CH₃OH, etc. Photocatalytic CO₂ conversion is largely achieved by different metal oxide-g-C₃N₄ based systems, as they have desirable band edges positions. Equations (4)–(7) are chemical reactions for CO₂ conversion to other solar fuels.





ZnO and TiO₂ are widely used as the g-C₃N₄-based composites for CO₂ conversion [429,430]. Wang et al. designed a photocatalyst comprising TiO₂ and g-C₃N₄ using ball milling and calcination. The heterostructure between TiO₂ and C₃N₄ leads to a low charge recombination rate, and high separation, resulting in the high CH₄ and CO evolution yields of 72.2 and 56.2 μmol g⁻¹ are obtained [431]. In another research, Nb-TiO₂/g-C₃N₄ Z-scheme heterojunctions were investigated and showed that the 50Nb-TiO₂/50g-C₃N₄ composition was the best photocatalysts with high carrier separation ability for the reduction of CO₂ [432]. The existence of electrons and holes in the CB of the g-C₃N₄ and VB of Nb-TiO₂, respectively, makes the Nb-TiO₂/g-C₃N₄ system a potential candidate for reducing of CO₂ into CH₄ and CO and HCOOH. Guo and coworkers were thermally deposited g-C₃N₄ onto the porous ZnO nanosheets by two-step calcination and demonstrated that the ZnO porous nanosheets @ g-C₃N₄-0.4 showed the highest CO₂ conversion efficiency [433]. Not only this composite suppressed the photoinduced electron recombination and facilitated the carrier transfer, but also the CO₂ chemisorption increased in this composite since the increasing defect vacancies formed on the porous ZnO nanosheets. Shen et al. demonstrated 3-ZnO/g-C₃N₄ (3 is the mass ratio of ZnO) has a high photocatalytic activity for CO₂ reduction to CO and CH₄ [434]. This experiment showed an insignificant decrease in photocatalytic activities, which indicates ZnO-g-C₃N₄ had high photocatalytic stability. A similar analysis has been performed on the hydrocarbon generation rate with hollow g-C₃N₄, hollow CeO₂, which is shown in Figure 18a [435]. The fast kinetics of CO reduction results in a higher CO evolution rate compared to the CH₄ and CH₃OH. The highest yield obtains for g-C₃N₄@ 49.7 wt % CeO₂. Increasing amounts of CeO₂ in the g-C₃N₄@CeO₂ composites will decrease the PL intensity at about 460 nm, meaning that the lower electrons and holes recombination and enhanced charge separation (Figure 18b) [435].

Other researchers also work on the CO₂ reduction of g-C₃N₄-metal oxide-based photocatalysts, which is listed in Table 3.

6.1.3. Photodegradation of Organic Pollutants

The development of an increased number of dye-related industries such as textile, food, and furniture manufacturing leads to severe environmental problems [445,446]. In addition to the negative aesthetic impact on water sources, the chemical oxygen demand (COD) in wastewater will be increased in the presence of organic dyes. Various methods, such as coagulation, adsorption, and membrane separation, have been used to eliminate organic dye from effluents, which only reclaim organic dyes from the wastewater liquid phase to the solid phase, creating secondary pollutants in the environment. These techniques are also a significant threat to living organisms. As a result, a metal oxide semiconductor has been widely used for the degradation of organic dyes. Photocatalysis in which most of the metal oxide can eliminate organic dyes by degradation and transfection them into particles, using solar energy for activation of the reaction. The related equations for dye degradation of the metal oxide-g-C₃N₄ are shown in Equations (8)–(11) [447]:

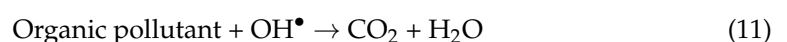
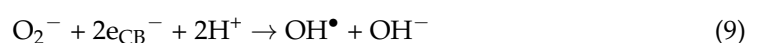


Table 2. A list of selected works on g-C₃N₄–metal oxide-based photocatalytic water splitting.

Photocatalyst	Type of Heterojunction	Source of Light	Highest Photocatalytic Rate	Ref
TiO ₂ -g-C ₃ N ₄	Type II	Asahi Spectra Hal-320 (300 mW cm ⁻²) with a 420 nm cut off filter ($\lambda > 420$ nm)	3.6 $\mu\text{mol}\cdot\text{h}^{-1}$	[115]
TiO ₂ -g-C ₃ N ₄	Type II	Xenon lamp with a 320 nm cut off filter ($\lambda > 320$ nm)	76.25 $\mu\text{mol}\cdot\text{h}^{-1}$	[414]
C-doped TiO ₂ -g-C ₃ N ₄	Type II	300 W Xe lamp (PLS-SXE300) with a 420 nm cutoff filter ($\lambda > 420$ nm)	35.6 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	[128]
TiO ₂ -g-C ₃ N ₄ decorated by Co-Pi	Type II	300 W Xe lamp coupled with a monochromator	-	
TiO ₂ nanodots/g-C ₃ N ₄	S-scheme	300 W Xe lamp (LANPU) with a 300 nm cutoff filter ($\lambda > 300$ nm)	The H ₂ and O ₂ evolution rate is 1318.3 and 638.7 $\mu\text{mol}\cdot\text{g}^{-1}$, respectively, (roughly as same as the stoichiometric ratio of evolved H ₂ to O ₂)	[415]
ZnO-g-C ₃ N ₄	Type II	PLS-SXE-300C lamp with an UV light intensity of 34 mW/cm ² and visible-light intensity of 158 mW/cm ²	-	[416]
N-doped ZnO-g-C ₃ N ₄	Z-scheme	PLS-SXE-300C UV lamp with a 420 nm cut off filter ($\lambda > 420$ nm)	152.7 $\mu\text{mol}\cdot\text{h}^{-1}$	[417]
g-C ₃ N ₄ -WO ₃	-	300-W Xe lamp (PLS-SXE300) with a 420 nm cutoff filter ($\lambda > 420$ nm)	963 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	[141]
O-g-C ₃ N ₄ /WO ₃	Z-scheme	300 W Xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	15,142 $\mu\text{mol}\cdot\text{g}^{-1}$	[265]
S-Cu ₂ O/g-C ₃ N ₄	Z-scheme	300 W Xe lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	24.83 $\mu\text{mol}\cdot\text{h}^{-1}$	[418]
BiO ₂ /g-C ₃ N ₄	Type II	500 W Xe lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	8,542 $\mu\text{mol}\cdot\text{g}^{-1}$	[419]
g-C ₃ N ₄ / BiYO ₃	Type II	-	37.6 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	[420]
g-C ₃ N ₄ /La _x Co _{3-x} O ₄	-	300 W Xe lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	63.12 $\mu\text{mol}\cdot\text{h}^{-1}$	[421]
Fe ₂ O ₃ /g-C ₃ N ₄	Z-scheme	350 W Xe lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	398.0 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	[210]
Mn ₃ O ₄ /g-C ₃ N ₄	p-n heterostructure	300 W Xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm) (PLS-SXE300D/300DUV, Beijing Perfectlight)	The H ₂ and O ₂ evolution rate is 3300 and 654 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$, respectively.	[422]
g-C ₃ N ₄ /Nitrogen-Doped Carbon Dots/WO ₃	-	300 W Xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm) (CEL-HXF 300)	3.27 mmol g ⁻¹ h ⁻¹	[423]
Mn ₃ O ₄ /g-C ₃ N ₄	-	300 W Xenon lamp source (PLS-SXE300D/300DUV)	2700 mmol·g ⁻¹ ·h ⁻¹	[422]
NiO/g-C ₃ N ₄	Type II	Xe lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	1.41 mmol·h ⁻¹	[424]
In ₂ O ₃ /g-C ₃ N ₄	Type II	300 W Xe lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	0.99 mmol·h ⁻¹	[425]
MoO _{3-x} -g-C ₃ N ₄	Z-scheme	300 W Xe lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	22.8 mmol·h ⁻¹	[426]
ZnO/Au/g-C ₃ N ₄	Z-scheme	150 W Xenon arc lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	3.69 $\mu\text{mol}\cdot\text{h}^{-1}\cdot\text{cm}^{-2}$	[397]
d-Ti ₃ C ₂ /TiO ₂ /g-C ₃ N ₄	-	300 W Xe lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	1.62 mmol·h ⁻¹ g ⁻¹	[396]
TiO ₂ /g-C ₃ N ₄	Type II	450 W high-pressure mercury lamp	22.4 mol·h ⁻¹	[402]
TiO ₂ -WO ₃ -g-C ₃ N ₄	-	-	286.6 mmol·h ⁻¹	[427]
TiO ₂ /Ti ₃ C ₂ /g-C ₃ N ₄	-	300 W Xe lamp	2592 mmol·g ⁻¹	[428]

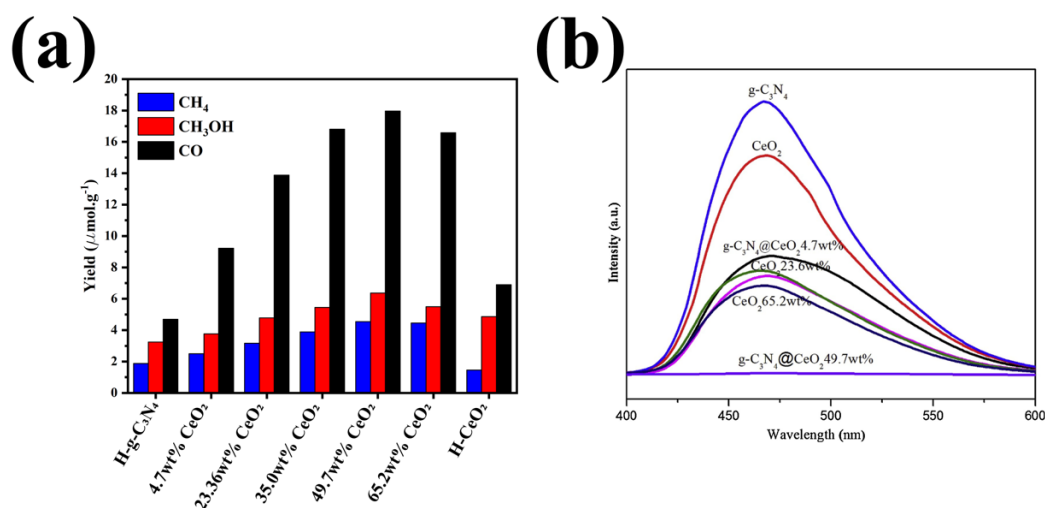


Figure 18. (a) Hydrocarbon generation of hollow g-C₃N₄ (H-g-C₃N₄), hollow CeO₂ (H-CeO₂), and g-C₃N₄@CeO₂ for 4 h illumination; (b) photoluminescence emission spectra of H-g-C₃N₄, H-CeO₂ references, and g-C₃N₄@CeO₂ heterojunction with different ratios. Copyright © 2022 Elsevier [435].

Researchers have suggested several metal oxide semiconductors that can be used in the g-C₃N₄-based heterojunctions. There are increasing numbers of studies showing the degradation capability of TiO₂, ZnO, WO₃, Bi₂O₃, CeO₂, etc. [100,101,113,116,448–451]. Zada et al. investigated the photodegrading of 2,4-dichlorophenol (2,4-DCP) and bisphenol A (BPA) over Au-(TiO₂/g-C₃N₄) nanocomposites. As mentioned above, the excellent photocatalytic of the nanocomposites containing Au is mainly due to the SPR of decorated Au [131]. The structure revealed 46% and 37% for 2,4-DCP and BPA degradation, which is 5.11 and 3.1 times larger than the bulk g-C₃N₄ in water under visible-light irradiation, respectively [131]. In another research, the synthesized ZnO@g-C₃N₄ exhibited an enhanced photocatalytic activity to degrade tetracycline (TC) under visible-light irradiation, which is 2.77 and 1.51 fold more than the photocatalytic ability of pure g-C₃N₄ and ZnO [452]. The improved degradation was due to the enhanced transference of charge carriers and reduced charge recombination in the presence of the generated reactive oxygen species (ROS). The pharmaceuticals contaminants have hazardous impacts on human health and environmental biodiversity. Zhu and coworkers fabricated WO₃-g-C₃N₄ composites for the photocatalytic degradation of one of the most well-known antibiotics, sulfamethoxazole (SMX), under visible light irradiation [453]. It is also revealed that the presence of RGO besides WO₃-g-C₃N₄ heterojunctions promoted the degradation rate of ciprofloxacin (CIP) nearly twice as compared to the WO₃-g-C₃N₄ structure [454]. In addition, H₂O₂ can raise the photocatalytic activities by the hydroxyl radicals' productions from the degradation of a natural organic matter up to 71% for 5 h [455]. Shafawi et al. prepared Bi₂O₃ particles decorated on porous g-C₃N₄ sheets by impregnation method [456]. 1 g/L of the composite containing g-C₃N₄ with 9 wt % Bi₂O₃ at 10 ppm reactive black 5 (RB 5) at pH = 5.7 demonstrated 84% degradation efficiency under UV-vis light for 120 min. The synergistic effects between g-C₃N₄ and CeO₂ provides higher catalytic activities compared to the bare ones [457–459]. The catalytic effects of the g-C₃N₄/CeO₂ composite, bare g-C₃N₄, and CeO₂ on the thermal decomposition of ammonium perchlorate (AP) were analyzed by using TGA and DTA characterization [460]. Two weight-loss regions from 25 °C to 500 °C, which were similar to the weight loss steps of AP in the absence of catalyst, were observed in Figure 19a,b. The weight loss decomposition temperatures of AP in the presence of the pure g-C₃N₄, CeO₂, and g-C₃N₄/CeO₂ were 53.6 °C, 47.6 °C and 74.6 °C, respectively (Figure 19a). It is also noticeable from Figure 19b that the AP thermal decomposition rate of g-C₃N₄/CeO₂ nanocomposites was higher than that of CeO₂ and g-C₃N₄. This heterojunction also shows a highly efficient for 2,4-dichlorophenol degradation [460].

Table 3. A list of CO₂ reduction applications of the g-C₃N₄-metal oxide-based photocatalytic.

Photocatalyst	Type of Heterojunction	Source of Light	Highest Photocatalytic Rate	Ref
NiO-g-C ₃ N ₄	Type II	300 W Xenon-arc lamp	4.17 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	[436]
g-C ₃ N ₄ foam-Cu ₂ O	Z-scheme	350–780 nm lamp	8.182 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$ (CO evolution)	[437]
NiMoO ₄ -g-C ₃ N ₄	Z-scheme	-	7238 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	[438]
CeO ₂ -g-C ₃ N ₄	Type II	300 W of Xenon-arc lamp	0.590 $\mu\text{mol}\cdot\text{h}^{-1}$ (CO evolution)	[439]
ZnO/Au/g-C ₃ N ₄	Z-scheme	300 W UV-Vis lamp	689.7 $\mu\text{mol}/\text{m}^2$ (CO evolution)	[440]
ZnO/g-C ₃ N ₄	Z-scheme	300 W xenon light source with a 420 nm cutoff filter ($\lambda > 420$ nm)	$\sim 72.24 \mu\text{mol}\cdot\text{g}^{-1}$	[435]
TiO ₂ /g-C ₃ N ₄	Type II	8 W UV lamp	The highest CH ₄ and CO yields of 72.2 and 56.2 $\mu\text{mol g}^{-1}$	[431]
Nb doped TiO ₂ /g-C ₃ N ₄	Z-scheme	1000 W Xe lamp	The CH ₄ , CO, O ₂ , HCOOH generation rate in the presence of 50Nb-TiO ₂ /50 g-C ₃ N ₄ is 562, 420, 1702, 698 $\mu\text{mol h}^{-1} \text{g}^{-1}$, respectively.	[432]
ZnO/g-C ₃ N ₄	Z-scheme	350 W Xe lamp	The CH ₃ OH production rate was 1.32 $\mu\text{mol h}^{-1} \text{g}^{-1}$	[429]
ZnO/g-C ₃ N ₄	Type II	300 W xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	H ₂ , CH ₄ , and CO production rates of 22.7 $\mu\text{mol}\cdot\text{g}\cdot\text{Cat}^{-1}\cdot\text{h}^{-1}$, 30.5 $\mu\text{mol}\cdot\text{g}\cdot\text{Cat}^{-1}\cdot\text{h}^{-1}$, and 16.8 $\mu\text{mol}\cdot\text{g}\cdot\text{Cat}^{-1}\cdot\text{h}^{-1}$	[433]
ZnO/g-C ₃ N ₄	Type II	350 W Xe arc lamp	45.6 $\text{mol}\cdot\text{g}\cdot\text{Cat}^{-1}\cdot\text{h}^{-1}$	[441]
TiO ₂ /g-C ₃ N ₄	Type II	450 W Xe lamp	22.5 $\mu\text{mol}\cdot\text{g}^{-1}$ and 70 $\mu\text{mol}\cdot\text{g}^{-1}$ for CO and CH ₄ yield, respectively	[442]
g-C ₃ N ₄ / 3D ordered microporous (3DOM)-WO ₃	Z-scheme	visible light ($\lambda \geq 420$ nm)	48.7 $\mu\text{mol g}^{-1} \text{h}^{-1}$	[443]
NiTO ₃ /g-C ₃ N ₄	Z-scheme	300 W xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	The highest yield of CH ₃ OH production is 13.74 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$	[444]

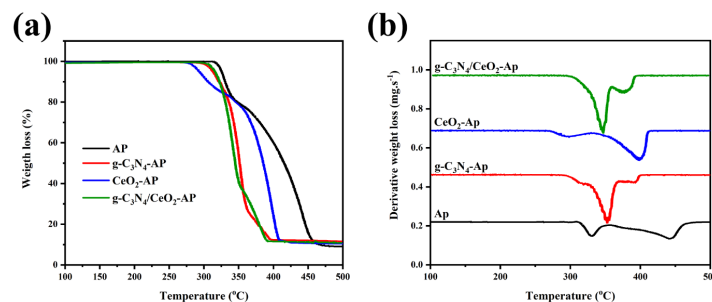


Figure 19. (a) TG and (b) DTG curves of pure ammonium perchlorate (AP), AP coupled with $g\text{-C}_3\text{N}_4$, CeO_2 , and $g\text{-C}_3\text{N}_4/\text{CeO}_2$ heterojunction, Copyright © 2022 Elsevier [460].

Methylene Blue (MB), Methylene Orange (MO), and Rhodamine B (RhB) are the most commonly stable dyes in water at room temperature [461–468]. MB and RhB are toxic dyes that their high concentration can be so harmful to human and marine animal health. In accordance with the high resistivity of the MB and RhB in different environmental conditions, wastewater treatment is an urgent issue. Therefore, it is crucial to provide an effective and low-cost system to eliminate MB and RhB from sewage. As mentioned above, 2D/1D $g\text{-C}_3\text{N}_4/\text{ZnO}$ nanocomposites reveal high stability, which retains the initial activity after repeated cycles [469]. The enhanced photocatalytic activity of $g\text{-C}_3\text{N}_4\text{-ZnO}$ is likely due to the synergistic effects of photon acquisition and direct contact between organic dyes and photocatalyst [470]. To be more specific, the RhB dye degradation efficiency of this composite is 99% and 95% after one and three cycles under sunlight irradiation, respectively [471]. It is reported that the highest degradation efficiency for MB is obtained for the 30 wt % few-layer $g\text{-C}_3\text{N}_4\text{-ZnO}$ nanocomposites is the best composition [472]. The higher amount of $g\text{-C}_3\text{N}_4$ can increase the electrons and holes recombination, leading to the decrease of photocatalytic activity. It is also noticeable that RhB degradation efficiency for this composite is about 2.1 times higher than that of pristine ZnO [473]. Apart from $g\text{-C}_3\text{N}_4\text{-ZnO}$ heterojunction, $g\text{-C}_3\text{N}_4\text{-TiO}_2$ heterojunction has high capability in the RhB degradation [474–477]. In these composites, $\bullet\text{O}_2^-$ played a major role while h^+ played a minor role [478–480]. Li et al. prepared Ti^{3+} self-doped TiO_2 nanoparticles/ $g\text{-C}_3\text{N}_4$ heterojunctions and demonstrated that the Ti^{3+} and O defects improve the conductivity and light absorption range to the visible wavelength region. Besides, the photocatalytic activities for environmental purification of organic compounds (MB) of the Ti^{3+} self-doped $\text{TiO}_2/g\text{-C}_3\text{N}_4$ nanostructures were improved under visible light irradiation remarkably [481]. It is proven that the photocatalytic activity of 2 wt % $g\text{-C}_3\text{N}_4\text{-TiO}_2$ improved by 70% compared to the bare TiO_2 [118]. The RhB degradation kinetic constant of $g\text{-C}_3\text{N}_4\text{-TiO}_2$ heterojunction is 9 times and 25 times higher than $g\text{-C}_3\text{N}_4$ and TiO_2 , respectively [482]. The $\text{AgPO}_4/g\text{-C}_3\text{N}_4$ was used for the degradation of organic pollutants. Z-scheme carbon nitride with AgPO_4 and Ag nanoparticles was used for the degradation of RhB. The presence of Ag can improve visible light absorption. The prepared hybrid structure can improve the electrons holes separation and efficiency [483]. Other metal oxides can also be used in the $g\text{-C}_3\text{N}_4$ based heterojunction for organic dye degradation [398,484–487]. $\text{Bi}_2\text{O}_3/g\text{-C}_3\text{N}_4$ heterojunctions are the most commonly used composites for organic dye degradation [488,489]. Fan et al. mentioned that the $\bullet\text{OH}$ radicals played the crucial roles during photocatalytic degradation of MB in 0.5- $\text{Bi}_2\text{O}_3/g\text{-C}_3\text{N}_4$ (0.5 is the mass of Bi_2O_3 in the synthetic process) Z-scheme heterojunction, while $\bullet\text{O}_2^-$, h^+ , e^- radicals have less contribution to this activity [490]. Compared to the pure Bi_2O_3 and bare $g\text{-C}_3\text{N}_4$, the $\text{Bi}_2\text{O}_3/g\text{-C}_3\text{N}_4$ heterojunctions showed better catalytic activity with 100% MB degradation ability in 90 min [490]. The 2% $\text{Bi}_2\text{O}_3/g\text{-C}_3\text{N}_4$ composites showed a degradation rate constant of 0.040 min^{-1} , which is 2.5 and 1.9 fold higher than that found for bulk and nitrogen vacant 2D $g\text{-C}_3\text{N}_4$ nanosheet, respectively [491]. This composition showed excellent photocatalytic activity with a methylene green degradation efficiency of 98.7% under visible light irradiation [448]. At 30 °C, 5% $\text{CeO}_2/g\text{-C}_3\text{N}_4$ (5% is the molar ratios of the $\text{CeO}_2/g\text{-C}_3\text{N}_4$ samples) photocatalyst showed

the best efficiency for MB degrading under visible light irradiation with the constant rate of 1.2686 min^{-1} , which is 7.8-fold higher than pure $g\text{-C}_3\text{N}_4$ [492]. Some ternary composites are also used for RhB and MB dye degradation [493]. $\text{SnO}_2\text{-ZnO}$ quantum dots anchored on $g\text{-C}_3\text{N}_4$ nanosheets were demonstrated as a promising candidate for RhB degradation, which is 99% in 60 min under visible-light irradiation [494]. This hybrid also showed a promising potential for hydrogen production with the photocatalytic rate of $13.61 \mu\text{mol g}^{-1}$, which is 1.06 and 2.27 times higher than that of the binary $\text{ZnO}/g\text{-C}_3\text{N}_4$ hybrid and pristine $g\text{-C}_3\text{N}_4$. In similar work, ZnS quantum dots ($\text{ZnS}/\text{SnO}_2/g\text{-C}_3\text{N}_4$) ternary nanocomposites were synthesized via solid-state calcination [495]. The high bandgap of SnO_2 and ZnS QDs leads to the reduction of the recombination rate and increases the separation of the generated carriers in the $g\text{-C}_3\text{N}_4$ [495]. The lower conduction band edge position of the SnO_2 , which is lower than others compel electrons to accumulate in this state. The position of the electrons and holes helped in improving the photocatalytic performance and enhancing stability. As a result, the transferred electrons can act as a suitable reductant and react with adsorbed O_2 to produce superoxide radicals (O_2^-). On the other side, the holes in the valence band of $g\text{-C}_3\text{N}_4$ can directly oxidize the pollutants to degraded products since these holes have strong oxidizing power [495]. $g\text{-C}_3\text{N}_4$ -metal oxide heterojunction is used to degrade different elements such as Hg, Cr (VI) [136,496,497], and different toxic and harmful materials such as atrazine (ATZ), chloramphenicol, ciprofloxacin (CIP), and etc. [197,454,498–502]. There are several research activities on the photodegradation applications of $g\text{-C}_3\text{N}_4$ -metal oxide-based heterojunctions which are listed in Table 4.

6.2. Sensors

Some benefits, such as high sensitivity to analytes, rapid response to external stimulations, excellent fluorescence quenching abilities, light and electricity conversion properties, biocompatibility, and high stability make $g\text{-C}_3\text{N}_4$ nanosheet a promising candidate as a modified electrode for sensors to detect different analytes such as glucose, hydrogen peroxide, dopamine, etc. [6,549–554]. Metal oxide semiconductors/ $g\text{-C}_3\text{N}_4$ composites are widely used as gas sensors [555]. As a result, the $g\text{-C}_3\text{N}_4$ loaded with metal oxides has also revealed new types of sensors to detect different kinds of materials [556]. General schematic description in Figure 20, shows different types of $g\text{-C}_3\text{N}_4$ -metal oxide sensors.

Like other applications, $g\text{-C}_3\text{N}_4\text{-TiO}_2$ based structures are one of the most used composites for sensing applications. Li et al. demonstrated that the photoelectrochemical $\text{TiO}_2/g\text{-C}_3\text{N}_4/\text{CdS}$ platform could be employed for the ultrasensitive determination of T4 polynucleotide kinase (T4 PNK) because this composite showed significant stability and reproducibility with high selectivity [557]. The photocurrent response is demonstrated in Figure 21A. To be more specific, “trace a” shows a low photocurrent at a bare electrode. The rapidly increased photocurrent is detected in “trace b” (from 10 to 30 s) due to decrease in the rate of electron-hole recombination and the high-efficiency absorption. In the $\text{TiO}_2/g\text{-C}_3\text{N}_4$ modified-FTO in “trace c”, the photocurrent increases to $30.7 \mu\text{A}$ since the bandgap of the $g\text{-C}_3\text{N}_4$ reduces the charge carrier recombination and facilitates the electron transmission [557]. The photocurrent increases in “trace d” ($\text{TiO}_2/g\text{-C}_3\text{N}_4/\text{CdS}$ nanocomposite-modified electrode). The photocurrent increased to $80.0 \mu\text{A}$ after attaching DNA_3 , which can capture ssDNA_2 to the electrode and then blocking 6-mercaptohexanol (MCH) (“trace e”). For comparison, the photocurrent of CdSe QDs with or without the bio-functionalization of DNA_2 on the electrode matrix is 180 and $78 \mu\text{A}$, respectively [557]. In addition, “trace f” shows that $\text{DNA}_2\text{-CdSe}$ QDs hybridized onto the surface, the photocurrent density increases to $180.0 \mu\text{A}$ since the presence of CdSe QDs improve the light absorption efficiency. To compare, CdSe QDs also functionalized the electrode matrix without DNA_2 , and the result showed that the photocurrent density is about $78.0 \mu\text{A}$ which is similar to the “trace e” and highly lower than “trace f”. As a result, the constructed platform shows a promising candidate for ultrasensitive detection of T4 PNK activity [557]. TiO_2 with $g\text{-C}_3\text{N}_4$ is also used for sensitive detection of protein kinase A (PKA), which is an enzyme that covalently decorates proteins with phosphate groups [558].

Table 4. the photodegradation application of g-C₃N₄-metal oxide-based heterojunctions.

Photocatalyst	Type of Heterojunction	Source of Light	Application	Highest Photocatalytic Rate	Stability	Ref
TiO ₂ -g-C ₃ N ₄	Z-scheme	300 W Xenon arc lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of Rhodamine B and tetracycline hydrochloride	The RhB removal rates for 5 layer TiO ₂ , 3, 5, 7 layers g-C ₃ N ₄ (0.5)/TiO ₂ were 5.1%, 17.9%, 31.2%, and 22.6%, respectively	-	[503]
P/O co-doped g-C ₃ N ₄ /anatase TiO ₂	Z-scheme	350 W Xenon-arc lamp as a light source with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of enrofloxacin	~98.5%	1 h	[504]
TiO ₂ @g-C ₃ N ₄	Z-scheme	100-W xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of RhB	95.68%		[505]
MoS ₂ -g-C ₃ N ₄ @TiO ₂	-	350 W Xenon lamp	Degradation of Methylene Blue	97.55	1 h	[506]
polyaniline-co-modified TiO ₂	-	xenon lamp containing an optical filter	Degradation of tetrabromobisphenol A	92.42%	16 h	[507]
N-TiO ₂ /O-doped N vacancy g-C ₃ N ₄	Type II Z-scheme	Lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of tetracycline hydrochloride and Cr(VI)	TC-HCl and Cr(VI) removal efficiency is 79.9% and 89.5%, respectively	-	[508]
TiO ₂ @g-C ₃ N ₄	Type II	300 W xenon lamp	Degradation of tetracycline antibiotic	The highest tetracycline degradation rate is 2.2 mg/min, which is 2 times higher than that of TiO ₂ and 2.3 times higher than that of bulk g-C ₃ N ₄ .	-	[509]
TiO ₂ /g-C ₃ N ₄ /persulfate (PS)	Type II	300 W xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of micropollutant (phenol, bisphenol A and carbamazepine)	99.3%.		[510]
TiO ₂ nanowire/g-C ₃ N ₄ nanosheet/graphene (G) heterostructures	-	300 W xenon lamp	Degradation of nitrobenzene	97%	4 h	[119]
Ti ³⁺ and O doped TiO ₂ /g-C ₃ N ₄	Type II	30 W cold visible light-emitting diode	Degradation of Rhodamine B	The photodegradation reaction rate constant based on this heterojunction is 0.0356 min ⁻¹ , which is 3.87 and 4.56 times higher than those of pristine Ti ³⁺ -TiO ₂ and g-C ₃ N ₄ , respectively.	-	[125]
TiO ₂ /g-C ₃ N ₄	Type II	-	Degradation of ciprofloxacin (CIP)	68.1%	3 h	[511]
ZnO-g-C ₃ N ₄	Z scheme	500 W Xe lamp, with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of Methylene Blue (MB)	75%	3 h	[512]
ZnO-g-C ₃ N ₄	Z scheme	300 W xenon lamp	Degradation of cephalixin oxidation	98.9%	1 h	[513]

Table 4. Cont.

Photocatalyst	Type of Heterojunction	Source of Light	Application	Highest Photocatalytic Rate	Stability	Ref
djembe-like ZnO-g-C ₃ N ₄	-	150 W Xenon light sources	Degradation of MB and RhB	MB and RhB degradation efficiency are ~95% and ~97%, respectively.	50 min	[514]
WO ₃ -g-C ₃ N ₄	-	300 W Xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of tetracycline	90.54%	1 h	[515]
WO ₃ -g-C ₃ N ₄	Z-scheme	450 W xenon lamps	Degradation of AO7	100%	75 min	[516]
WO ₃ -g-C ₃ N ₄	Z-scheme	35-W Xe lamp with a radiation intensity of 1380 $\mu\text{W}\cdot\text{cm}^2$	Degradation of orange G	98%	1 h	[517]
Ag-WO ₃ /g-C ₃ N ₄	Z-scheme	500 W Xe lamp (Beijing Bofei Technology. Co. Ltd., Beijing, China)	degradation of oxytetracycline hydrochloride	97.74	1 h	[273]
WO ₃ @g-C ₃ N ₄ @MWCNT	Z-scheme	5 mW cm^{-2} Xe lamp (SANEI electronics-JAPAN)	Degradation of tetracycline	79.54%	2 h	[518]
g-C ₃ N ₄ /WO ₃	Z-scheme	-	Degradation of nitenpyram	The photocatalyst rate constant is 0.036 min^{-1} which is about 1.7 and 25 times higher than that of pure g-C ₃ N ₄ and WO ₃ , respectively.	-	[519]
Bi ₂ O ₃ -g-C ₃ N ₄	Z-scheme	350 W xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of Rhodamine B	98%	80 min	[520]
Bi ₂ O ₃ -g-C ₃ N ₄	Z-scheme	250 W Xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of tetracycline	80.2%	50 min	[521]
g-C ₃ N ₄ -CeO ₂	Type II	300W Xe arc lamp	Degradation of antibiotic doxycycline hydrochloride	84%	1 h	[522]
Shuttle-like g-C ₃ N ₄ -CeO ₂	-	300 W Xe arc lamp	Degradation of norfloxacin	88.6%	1 h	[523]
Kaolin/CeO ₂ /g-C ₃ N ₄	-	500 W Xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Removal of ciprofloxacin	90%	150 min	[524]
Fe ₃ O ₄ /CeO ₂ /g-C ₃ N ₄	-	300 W Xe lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of tetracycline hydrochloride	96.63%	3 h	[525]
Au/g-C ₃ N ₄ nanosheets/CeO ₂	Z-scheme	500 W Xe lamp with a 400 nm cutoff filter ($\lambda > 400$ nm)	Reduction of hexavalent chromium Oxidation of oxytetracycline hydrochloride	88.2% 95.1%	150 min	[411]
Co ₃ O ₄ -g-C ₃ N ₄	-	250 W Xe lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of Methyl Orange	100%	3 h	[526]

Table 4. Cont.

Photocatalyst	Type of Heterojunction	Source of Light	Application	Highest Photocatalytic Rate	Stability	Ref
NiO-g-C ₃ N ₄	Type II	500 W Xe-lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of Methylene Blue	6.3 wt. % NiO loading shows a 2.3 times higher MB degradation rate than that of the pristine g-C ₃ N ₄ .	80 min	[527]
V ₂ O ₅ -g-C ₃ N ₄	Z-scheme	500 W Xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of Congo Red and Cr (VI) reduction	-	90 min	[528]
MoO ₃ -g-C ₃ N ₄	Z-scheme	150 W-Xe lamp having 1.5 AM filter which allows wavelength larger than 400 nm for the visible light-based catalytic reaction	Degradation of Rhodamine B	93%	3 h	[529]
MoO ₃ -g-C ₃ N ₄	Z-scheme	500W Xenon lamp	Degradation of Rhodamine B	100%	10–15 min	[530]
MoO ₃ -g-C ₃ N ₄	Z-scheme	visible light ($\lambda > 420$ nm)	Degradation of tetracycline	85.9%	100 min	[531]
BiMoO ₆ -g-C ₃ N ₄	Z-scheme	300 W Xe lamp	Degradation of ciprofloxacin	~100	30 min	[532]
g-C ₃ N ₄ /SnO ₂	S-scheme	A 300 W Osram, 230 V with a 420 nm cut-off filter as used as the visible light source.	Degradation of NO	44.17%	30 min	[533]
g-C ₃ N ₄ -NiO	Z-scheme	30 W LED-light source	Degradation of Methyl Orange (MO)	96.8%	2 h	[461]
ZnO/g-C ₃ N ₄	Type II	150 W Xe lamp	Degradation of MB and RhB	The MB and RhB degradation was ~95% and ~97%, respectively.	50 min	[514]
g-C ₃ N ₄ /CuO _x	-	350W Xe lamp	Degradation of Methyl Orange (MO)	~62.5%.	70 min	[534]
WO ₃ /g-C ₃ N ₄	Z-scheme	300 W Xe arc lamp	Degradation of sulfamethoxazole	91.7%	4 h	[453]
g-C ₃ N ₄ Nanosheets/ZnO	-	500 W Xe lamp	Photocatalytic reduction of aqueous chromium(VI)	70%	4 h	[535]
ZnO/g-C ₃ N ₄	-	500 W xenon lamp	Photodegradation of Direct Blue 199 (DB)	99%	100 min	[536]
ZnO/g-C ₃ N ₄	-	A 150 W xenon lamp	Degradation of tetracycline hydrochloride	97%	30 min	[452]
MoS ₂ /Al ₂ O ₃ /g-C ₃ N ₄	-	150 W tungsten halogen lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of crystal violet (CV)	97.3%.	90 min	[537]
g-C ₃ N ₄ /ZnO	Z-scheme	300 W Xenon lamp cutoff filter with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of 4-chlorophenol	~ 95%	1 h	[538]

Table 4. Cont.

Photocatalyst	Type of Heterojunction	Source of Light	Application	Highest Photocatalytic Rate	Stability	Ref
Gd ₂ O ₃ NPs@g-C ₃ N ₄	-	300 W Xe lamp with a 420 nm cutoff filter ($\lambda > 420$ nm) (PLS-SXE300, Beijing Perfectlight Technology Co., Ltd., Beijing, China)	Degradation of Methyl Orange (MO) Methyl Blue (MB) Rhodamine B	72.4% 95.5% 100%	2 h	[539]
MoO ₃ /g-C ₃ N ₄ /peroxydisulfate (PDS)	Z-scheme	350 W Xenon lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	degradation of ofloxacin (OFLX)	94.4%	2 h	[540]
ZnO/g-C ₃ N ₄	Z-scheme	300 W Xe lamp	Degradation of cephalixin	98.9%	1 h	[513]
Ce ₂ O/Bi ₂ O ₃ /g-C ₃ N ₄	Z-scheme	75 W halogen lamp	Degradation of Malachite green and Rose Bengal	-	1 h	[541]
CuO/ZnO/g-C ₃ N ₄	Z-scheme	400 W hallow lamp	Degradation of Methylene Blue and ammonia-nitrogen	The MB and ammonia-nitrogen degradation efficiency are ~98% in 45 min and 91% in 6 h.	45 min, and 6 h	[542]
Fe ₃ O ₄ /ZnO/g-C ₃ N ₄	Type II	23 W white LED	Degradation of pantoprazole	97.09%.	90 min	[543]
Fe ₃ O ₄ /TiO ₂ /g-C ₃ N ₄	-	500 W Xenon Lamp with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of Rhodamine B (RhB) and Methylene Orange (MO)	The photocatalyst shows the RhB, and MO degradation efficiency is ~96.4% in 80 min and 90% in 120 min.	8 min and 120 min	[544]
g-C ₃ N ₄ /ZnO/TiO ₂	-	1000 W xenon lamp irradiation system equipped with a 410 nm cutoff filter under room temperature.	Degradation of p-toluenesulfonicacid (p-TSA)	100%	60 min	[545]
ZnO/Ag ₂ O/g-C ₃ N ₄	-	high pressure xenon short arc lamp with the light intensity of 100 mW cm ⁻²	Degradation of ciprofloxacin	97.4%	48 min	[196]
WO ₃ /TiO ₂ @g-C ₃ N ₄	-	500 W metal halide lamp	Removal of acetylsalicylate (aspirin) and methyl-theobromine (caffeine)	98%	90 min	[546]
TiO ₂ @g-C ₃ N ₄ /Co ₃ O ₄	-	300 W xenon lamp	Degradation of tetracycline (TC) and Methylene Orange (MO)	The TC (10 mg/L) and MO (25 mg/L) degradation efficiency are 91.6% and 97.8%, respectively.	1 h	[547]
g-C ₃ N ₄ /Bi ₂ O ₃ /TiO ₂	-	Xe-lamp light source with a 420 nm cutoff filter ($\lambda > 420$ nm)	Degradation of Methylene Blue (MB)	The MB removal efficiency is 77.5%.	3 h	[548]
SnO ₂ /ZnO@g-C ₃ N ₄	-	300 W xenon lamp	Degradation of Rhodamine B dye and H ₂ production	99%	1 h	[494]
g-C ₃ N ₄ /NiO/ZnO/Fe ₃ O ₄	-	-	removal of esomeprazole	95.05 ± 1.72%	70 min	[513]

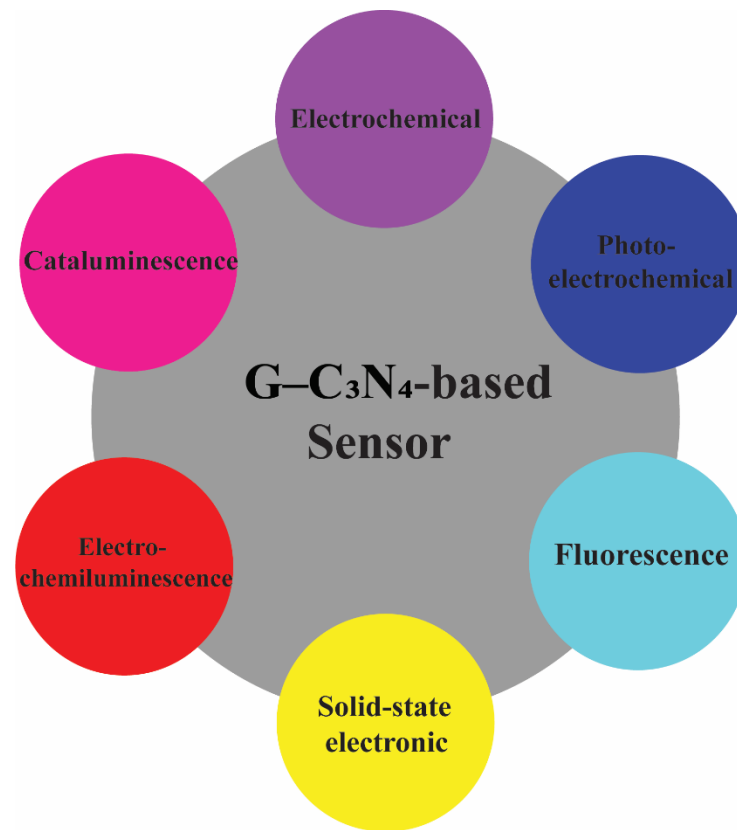


Figure 20. Different kinds of $g\text{-C}_3\text{N}_4$ -metal oxide-based sensors for materials detection.

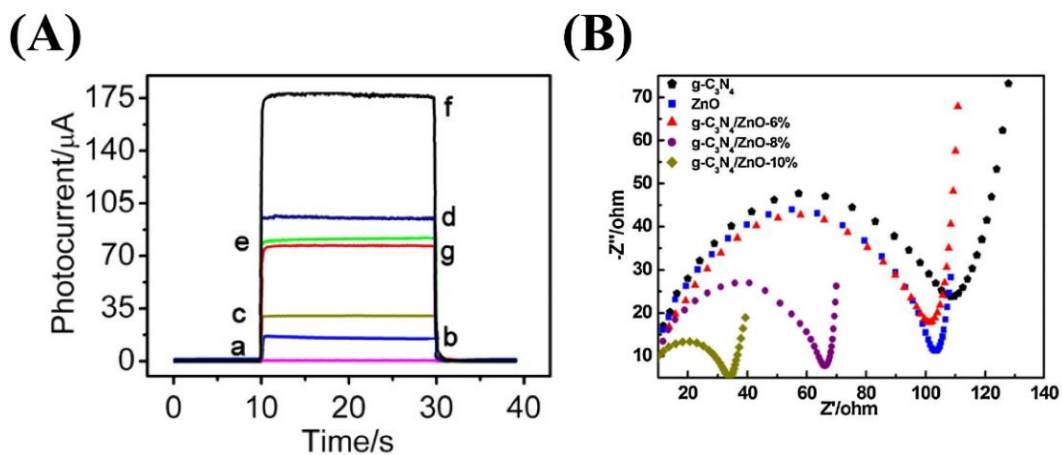


Figure 21. (A) Photocurrent responses of (a) FTO, (b) FTO/TiO₂, (c) FTO/TiO₂/ $g\text{-C}_3\text{N}_4$, (d) FTO/TiO₂/ $g\text{-C}_3\text{N}_4$ /CdS, (e) FTO/TiO₂/ $g\text{-C}_3\text{N}_4$ /CdS/capture-DNA₃/MCH, (f) FTO/TiO₂/ $g\text{-C}_3\text{N}_4$ /CdS/capture-DNA₃/MCH/DNA₂-CdSe, (g) FTO/TiO₂/ $g\text{-C}_3\text{N}_4$ /CdS/capture-DNA₃/MCH/CdSe, Copyright © 2022 American Chemical Society [557]; (B) Nyquist plots of ZnO, $g\text{-C}_3\text{N}_4$ /ZnO composites and $g\text{-C}_3\text{N}_4$ nanosheets electrodes, Copyright © 2022 American Chemical Society [559].

ZnO- $g\text{-C}_3\text{N}_4$ is another heterojunction used as an agent as UV-assisted gas sensors. It is shown that the ethanol ($\text{C}_2\text{H}_5\text{OH}$) sensing capability of ZnO- $g\text{-C}_3\text{N}_4$ is much higher when compared to the bare ZnO and $g\text{-C}_3\text{N}_4$. The ZnO with 8% $g\text{-C}_3\text{N}_4$ showed the best sensing performance than the others, which attributes to effective electrons and holes separation between $g\text{-C}_3\text{N}_4$ and ZnO and the UV-light catalytic effect in the room temperature [559]. The structure showed an excellent response of ethanol at room temperature, which was

higher than the pure ZnO at the same condition. The arc radius of ZnO and g-C₃N₄/ZnO is shown in Figure 21B. The results show that adding g-C₃N₄ nanosheets to ZnO improves the charge transfer and reduces the electrical resistivity [559].

Sensing of NO₂ and CH₄ by ZnO-g-C₃N₄ heterojunction have also been investigated [560,561]. Hydrogen sulfide (H₂S) is a corrosive and toxic gas, which can be generated from oil refining, mines, or petroleum fields, fuel cells, or food processing industries and may cause severe problems. As a result, eliminating H₂S has always been a serious research topic. Zeng et al. used α -Fe₂O₃/g-C₃N₄ for H₂S detection and showed that 5.97% α -Fe₂O₃ with g-C₃N₄ provides the best cataluminescence response [562]. The prepared α -Fe₂O₃/g-C₃N₄ composite can be used as an H₂S gas sensor in environmental monitoring, oil refining, food processing, and so forth. Researchers have shown that g-C₃N₄-SnO₂ composites can also be used as sensors for different kinds of compounds. Specifically, Zhang et al. proved that the presence of g-C₃N₄ beside SnO₂ enhances the gas sensitivity and selectivity of SnO₂ to acetic acid vapor with the reduced temperature from 230 °C to 185 °C [563]. The limit of acetic acid vapor detection is 0.1 ppm; however, the long-term stability of the prepared composite is low and should be improved. In-SnO₂ loaded cubic mesoporous g-C₃N₄ will be a new method and structure to design efficient humidity sensors for monitoring indoor climatic conditions [564]. Table 5 lists the applications of g-C₃N₄-metal oxide heterojunction used as sensor material.

6.3. Bacterial Disinfection

According to different sizes, shapes, and structures of bacteria, viruses, microbes, and microalgae, the g-C₃N₄ has the ability to eliminate them under ultraviolet or visible light irradiation changes. Fabricating the g-C₃N₄ with different metal oxides is important for effective composites for water antimicrobial disinfection and microbial control. Bio-hazards are widely present in wastewater and contaminated water containing a variety of viruses, bacteria, fungi, etc., causing health issues in humans [30]. The pioneering work by Matsunaga et al. showed that TiO₂ could help in inactivating bacteria such as *Escherichia coli*, *Lactobacillus acidophilus*, and *Saccharomyces cerevisiae* under UV light [580]. TiO₂ suffers from low absorption of solar energy and this is due to the wide bandgap of TiO₂, which is widely discussed in the previous parts. One method to increase TiO₂ performance is to combine it with different semiconductors. Li et al. prepared g-C₃N₄-TiO₂ using a facile hydrothermal-calcination approach with high photocatalytic bacterial inactivation ability [114]. They demonstrated that this composite could facilitate water disinfection, especially in hospital wastewaters with highly concentrated pathogenic microorganisms, using visible light. In another work, researchers investigated the excellent efficiency of this composite on the removal of the microcystis aeruginosa and Microcystin-LR [581].

In order to improve the photocatalytic activity, researchers are more likely to dope metal elements such as Ag and Cr in the g-C₃N₄-metal oxide structure, which can perform as an electron sink to separate the carriers. Doping these elements not only can modify the band edge positions of these structures but also, due to antimicrobial and bacterial properties of these materials, they are widely used in the g-C₃N₄-metal oxide-based photocatalysts [102,189]. g-C₃N₄/Cr-ZnO nanocomposites with superior antibacterial activity against Gram-negative (*Escherichia coli*) and Gram-positive (*Bacillus subtilis*, *Staphylococcus aureus*, and *Streptococcus salivarius*) were investigated [172]. In this research, the 60%g-C₃N₄/5%Cr-ZnO nanocomposite performs the highest antibacterial activity. Fe-SnO₂/g-C₃N₄ revealed a promising sterilization performance for *Escherichia coli* and *Staphylococcus aureus* under sunlight, near-ultraviolet light, and daylight lamp [582]. The sterilization performance of this structure is mostly deserved under a daylight lamp. The magnetic silver-iron oxide nanoparticles decorated graphitic carbon nitride nanosheets showed antibacterial performance against *E. coli* bacteria [583].

Table 5. The list of applications of g-C₃N₄–metal oxide heterojunction used for detection.

Photocatalyst	Application	Explanation	Ref
MoO ₃ -g-C ₃ N ₄	Detection of Furazolidone	The high electroactive surface area (0.3788 cm ²), as well as enhanced heterogeneous electron transfer rate ($K^{\circ}_{\text{eff}} = 4.91 \times 10^{-2} \text{ cm} \cdot \text{s}^{-1}$) can detect Furazolidone with low limit of detection (LOD) (1.4 nM) with a working range of 0.01–228 μM.	[528]
Co ₃ O ₄ -g-C ₃ N ₄	Detection of environmental phenolic hormones	This composite showed a wide detection range and a low limit of detection LOD (10 ⁻⁹ mol L ⁻¹)	[565]
g-C ₃ N ₄ -Fe ₃ O ₄	Determination of Tramadol in Human Biological Fluids	LOD of this composite is ~0.1 μM	[566]
V ₂ O ₅ -g-C ₃ N ₄	Detection of folic acid	The sensitivity, the LOD, and noise-to-signal ratio of the sensor is 19.02 μA mM ⁻¹ cm ⁻² , and 0.00174 μM, and 3 (S/N = 3), respectively	[567]
g-C ₃ N ₄ -NiO	Detection of quercetin	The dynamic range and LOD of g-C ₃ N ₄ -NiO for sensing quercetin is 10 nM to 250 and 0.002 μM, respectively	[568]
Cu ₂ O-g-C ₃ N ₄	Humidity sensor	The response time and recovery time were 180–200 s and 5–10 s, respectively.	[569]
NiO-Co ₃ O ₄ -g-C ₃ N ₄	Detection of tetrabromobisphenol-A	This structure showed a LOD of ~0.1 mmol L ⁻¹	[570]
ZnO @ g-C ₃ N ₄	Detection of CCRF-CEM cells	The LOD of this composite is ~20 cell/mL	[571]
ZnO flower-rod/g-C ₃ N ₄ -gold nanoparticle	Detection of carcinoembryonic antigen (CEA)	The PEC aptasensor for CEA determination is from 0.01 to 2.5 ng·mL ⁻¹ with detection of 1.9 pg·mL ⁻¹ .	[572]
g-C ₃ N ₄ /ZnO	Ethanol sensing	Compared to the ZnO, the g-C ₃ N ₄ /ZnO-8% composites revealed an excellent response ~60 orders of magnitude at room temperature.	[559]
ZnO/g-C ₃ N ₄	NO ₂ sensing	The response, recovery time, and LOD of ZnO/g-C ₃ N ₄ -10 wt % are 142, 190 s, and 38 ppb, respectively.	[561]
SnO ₂ /g-C ₃ N ₄	Ethanol gas Sensing	The composite with 7 wt % g-C ₃ N ₄ content exhibited a promising gas sensing property to ethanol, which has better response and selectivity than that of the pure SnO ₂ based sensor.	[321]
g-C ₃ N ₄ /ZnO	CH ₄ Sensing	The higher active sites can be obtained in this structure due to the larger specific surface area leading to the great response toward 1000 ppm CH ₄ .	[560]
g-C ₃ N ₄ -Mn ₃ O ₄	H ₂ S sensor	The LOD (S/N = 3, LOD) is 0.13 μg mL ⁻¹	[573]
α-Fe ₂ O ₃ /g-C ₃ N ₄	H ₂ S sensor	The linear detection range and detection limit of the H ₂ S gas sensor were 0.88–7.01 μg mL ⁻¹ (r = 0.998) and 0.5 μg mL ⁻¹ (S/N = 3), respectively.	[562]
O vacancy WO _{2.9} /g-C ₃ N ₄	4-nitrophenol Sensing	Compared to other research works, this heterojunction showed the linear range of 0.4–100 μmol/L and a lower detection limit of 0.133 μmol/L.	[574]
b-Bi ₂ O ₃ /g-C ₃ N ₄	Detection of chlorpyrifos	The linear detection range and detection limit of this sensor were 0.1–80 ng mL ⁻¹ a03 μg mL ⁻¹ , respectively.	[575]
Fe ₃ O ₄ /Bi ₂ O ₃ /g-C ₃ N ₄	Determination of Cd ²⁺ and Pb ²⁺	The minimum quantity for Cd ²⁺ and Pb ²⁺ that can be detected is 3 × 10 ⁻⁹ and 1 × 10 ⁻⁹ mol/L, respectively.	[576]
WO ₃ /g-C ₃ N ₄	Detection of phosmet	The limit of detection (LOD) and limit of quantification (LOQ) of this device was calculated 3.6 nM and 11.2 nM, respectively.	[577]
WO ₃ /g-C ₃ N ₄ /MnO ₂	Detection of oxytetracycline cathodic	This sensor exhibited a wide detection range from 1 pM to 150 nM and a low detection limit of 0.1 pM.	[578]
CuO-g-C ₃ N ₄	Aflatoxin B1 sensing	The limit of detection of 6.8 pg mL ⁻¹ for AFB1.	[579]

Zhao and coworkers illustrated that g-C₃N₄/ZnO/cellulose (CNZCel) shows the high thermal stability of the composite and photo-excited carriers separation efficiency and decreases the recombination rate [202]. Besides, the nanocomposite revealed excellent antibacterial activity against *Escherichia coli* (*E. coli*) and Gram-positive bacteria *Staphylococcus aureus* (*S. aureus*). The presence of ZnO in the structure can efficiently enhance the antibacterial activities against *E. coli* and *S. aureus*. Compared to the other materials, these ternary composites demonstrated better antibacterial performance (Figure 22a,b). The effect of the g-C₃N₄ content on the antibacterial property against *E. coli* and *S. aureus* in this composite is also investigated, which is shown in Figure 22c,d [202]. The mechanism of antibacterial activity generates electron-hole pairs under illumination, which can provide enormous reactive oxygen species (ROS), such as $\bullet\text{O}_2^-$, $\bullet\text{OH}_2$ and H_2O_2 , needed for the inhibition of *E. coli* and *S. aureus* growth.

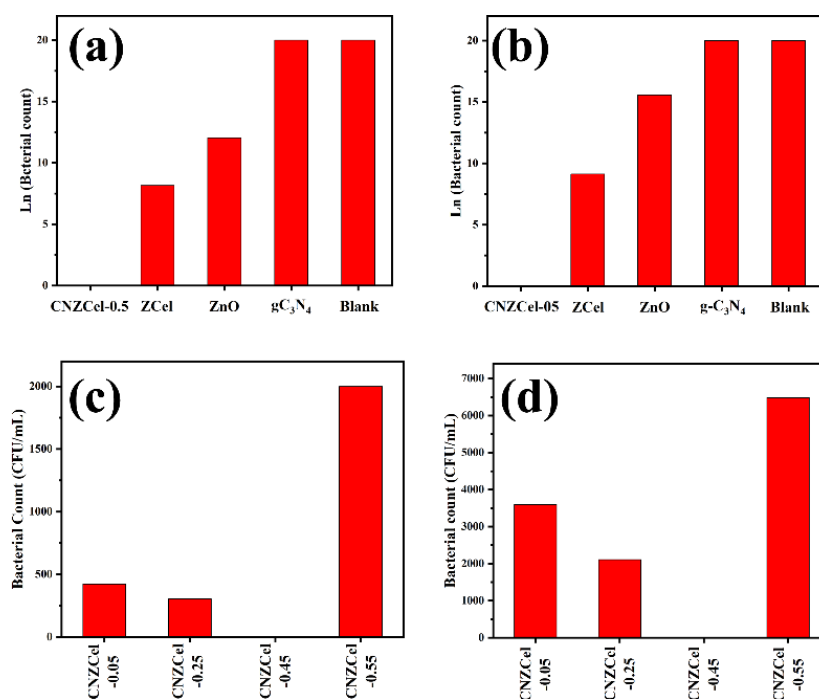


Figure 22. (a) Plots of bacterial counts of g-C₃N₄/ZnO/cellulose-0.45 (0.45 corresponds to the mass of g-C₃N₄ addition), ZnO/cellulose, ZnO, g-C₃N₄, and blank sample against *S. aureus* and (b) *E. coli*; (c) bacterial counts of g-C₃N₄/ZnO/cellulose composites against *S. aureus* and (d) *E. coli*. Reproduced from Reference [202].

Table 6 mentioned some additional studies on the disinfection ability of g-C₃N₄-metal oxide-based nanomaterials.

Table 6. Number of researches on the disinfection ability of the g-C₃N₄ metal oxide-based nanomaterials.

Photocatalyst	Source of Light	Application	Highest Photocatalytic Rate	Ref
g-C ₃ N ₄ /TiO ₂ /Ag	Xe lamp with a 420 nm cutoff filter (λ > 420 nm)	Bactericidal efficiency against <i>E. coli</i>	the optimal bacterial inhibition of g-C ₃ N ₄ /TiO ₂ /Ag was 84%	[102]
g-C ₃ N ₄ /Cu ₂ O	300 W xenon lamp with 400 nm cutoff filter (λ > 400 nm)	Inactivation efficiencies of <i>E. coli</i> as well as <i>Fusarium graminearum</i>	g-C ₃ N ₄ with 45%Cu ₂ O composition revealed the inactivation efficiencies of ~7 log <i>E. coli</i>	[584]
TiO ₂ /g-C ₃ N ₄ /SnO ₂	300 W xenon lamp with 420 nm cutoff filter (λ > 420 nm)	Bactericidal efficiency against <i>E. coli</i>	TiO ₂ /g-C ₃ N ₄ /SnO ₂ structure showed a good <i>E. coli</i> disinfection efficiency under visible and UV irradiation, with ~−6.7 log <i>E. coli</i> and −8.2 log <i>E. coli</i> , respectively.	[314]
α-Fe ₂ O ₃ /CeO ₂ decorated g-C ₃ N ₄	500 W Xe light with the cut-off filter of ~420 nm (λ > 420 nm)	Antibacterial activities against <i>S. aureus</i> (G+) and <i>E. coli</i> (G−) bacteria	α-Fe ₂ O ₃ /CeO ₂ decorated g-C ₃ N ₄ exhibited perfect antibacterial to <i>E. coli</i> and <i>S. aureus</i> activity with the maximum zone of inhibition (ZOI) of 11 ± 0.5, 12 ± 0.	[585]
g-C ₃ N ₄ -m-Bi ₂ O ₄	500 W halogen lamp with UV-cut off filter (λ > 420 nm)	Bactericidal efficiency <i>E. coli</i> and <i>S. aureus</i> bacteria	The ZOI value for <i>E. coli</i> and <i>S. aureus</i> bacterial strains was ~11±0.5 mm and 12–13 ± 0.5.	[586]
Ag/ZnO/g-C ₃ N ₄	300 W xenon arc lamp	Bactericidal efficiency against <i>E. coli</i>	The <i>E. coli</i> disinfection efficiency of Ag/ZnO/g-C ₃ N ₄ structure is ~7.4 log <i>E. coli</i> .	[190]
Ag/AgO-g-C ₃ N ₄	100 W tungsten lamp	Bactericidal efficiency against <i>E. coli</i>	The 1, 2, 3, and 4 mg of catalyst showed low quantitative <i>E. coli</i> growth inhibition, which was ~17% and 65%, 97%, 99%, respectively.	[587]
Cu ₂ O-g-C ₃ N ₄	36 W fluorescent lamp with the cut-off filter of ~400 nm (λ > 400 nm)	Bactericidal efficiency against <i>B. subtilis</i> , <i>E. coli</i> , <i>S. aureus</i> and <i>P. aeruginosa</i>	The maximum ZOI for Cu ₂ O-g-C ₃ N ₄ to <i>B. subtilis</i> , <i>E. coli</i> , <i>S. aureus</i> and <i>P. aeruginosa</i> is 22 ± 1.67, 15 ± 1.08, 11 ± 1.22, 6 ± 0.09, respectively.	[587]
g-C ₃ N ₄ / TiO ₂ Kaolinite	Xenon lamp with a 400 nm cut-off filter	Disinfection ability towards <i>S. aureus</i>	The disinfection efficiency of g-C ₃ N ₄ /TiO ₂ /kaolinite is ~4.3 log cfu/mL in 5 h.	[588]
TiO ₂ /g-C ₃ N ₄	500 W xenon arc lamp with a 400 nm cut-off filter	Anti-fouling ability of <i>E. coli</i>	TiO ₂ /g-C ₃ N ₄ showed an excellent <i>E. coli</i> removal with a permeate flux of 2 times higher than that of filtration alone.	[589]
TiO ₂ /g-C ₃ N ₄	-	Bactericidal efficiency against <i>E. coli</i>	The bacterial survival rate for the TiO ₂ nanotube/g-C ₃ N ₄ nanofilms is ~16%.	[590]
g-C ₃ N ₄ /Ag-TiO ₂	Xenon lamp	Bactericidal efficiency against both Gram-negative 18 <i>Escherichia coli</i> and Gram-positive <i>Staphylococcus aureus</i>	The presence of Ag in g-C ₃ N ₄ /Ag-TiO ₂ structure could enhance water disinfection under visible light.	[591]
g-C ₃ N ₄ /MoS ₂ -Bi ₂ O ₃	300 W xenon arc lamp	Bactericidal efficiency against <i>E. coli</i>	g-C ₃ N ₄ /Bi ₂ O ₃ with the ratio of 1:0.5, could entirely inactivate 6-log ₁₀ cfu/mL <i>E. coli</i> .	[586]

6.4. Other Applications

Due to the specific properties of g-C₃N₄-based heterojunctions, these composites can be used in many other applications [150,247,585,592]. With reference to the high energy demand for electronic devices and vehicles, rechargeable lithium-ion batteries (LIBs) have captured enormous attention among many researchers [319,320,593–595]. Anode materials, which are a significant part of the LIBs, should have improved specific capacity, high stability. SnO₂ is a new type of lithiophilic material with high specific capacity (1494 mAh g⁻¹), low potential for Li⁺ insertion, increased number of sources, and etc. [320,593,596–602]; however, it suffers from significant volume expansion (~300%) during the charge-discharge cycling, leading to fast capacity fading. SnO₂ nanosheets with 20–25 nm thickness dispersed in the g-C₃N₄ showing the potential lithium storage for LIBs. SnO₂@C₃N₄ nanocomposites can also be prepared by a scalable solid-state reaction [319]. Tran et al. used the hydrothermal method to grow SnO₂ onto the graphite oxide/g-C₃N₄ [320]. This composite showed an excellent reversible capacity and cycling performance for lithium storage, which may attribute to the existence of g-C₃N₄ or graphite oxide-g-C₃N₄. SnO₂@g-C₃N₄ based nanocomposites provide a suitable substitute for next-generation high-power and low-cost LIBs [319]. Zn₂GeO₄ nanoparticles demonstrate high capacity and act as spacers to prevent the g-C₃N₄ sheets from stacking, leading to the expanded interlayer and exposed vacancies for higher Li-ion storage. Besides, g-C₃N₄ layers, in turn, reduce the expansion of the particles and provide more stable solid electrolyte interphase, results in highly reversible lithium storage capacity, which is 1370 mA h g⁻¹ at 200 mA g⁻¹ after 140 cycles with a significant rate capability of 950 mA h g⁻¹ at 2000 mA g⁻¹ [595]. The SnO₂ nanosheets with g-C₃N₄ can enhance lithium storage capabilities and cycling performance [593]. Other g-C₃N₄-metal oxide nanocomposites can also be used in the supercapacitor that can be used with batteries to mitigate the power delivery problems associated with batteries [596–598]. The bare g-C₃N₄, g-C₃N₄/CuO, g-C₃N₄/Co₃O₄ electrode-based device exhibited a specific capacitance of 72 F g⁻¹, 95 F g⁻¹, and 201 F g⁻¹, respectively [599]. Besides, the energy density of g-C₃N₄/CuO and g-C₃N₄/Co₃O₄ at the constant power density of 1 kW·kg⁻¹ are 13.2 W·h·kg⁻¹, and 27.9 W·h·kg⁻¹, respectively. Moreover, the excellent theoretical capacities of V₂O₅ for the next generation supercapacitors makes the porous g-C₃N₄@V₂O₅ good candidate with a high specific capacity of about 457 Fg⁻¹ at 0.5 Ag⁻¹ with high cycling performance (~84% after 500 cycles) [600].

Photocatalytic nitrogen fixation, which is a clean and sustainable method for the production of NH₃, is another application of g-C₃N₄-based materials. However, insignificant surface-active sites, poor stability, and high recombination rate of carriers have restricted the efficiency of g-C₃N₄ for nitrogen fixation activities. The carrier separation should be enhanced by doping with other elements or compositing with various metal oxides. Finally, nitrogen gas should completely adsorb on the photocatalyst for the reduction steps [603]. It is shown that the cyano group (–C≡NC) in cyano group modified g-C₃N₄ enhance the photocatalytic applications for N₂ fixation up to 128 times [604].

As we discussed earlier, iodine and sulfur are atoms that can improve the carriers' activities in the bulk g-C₃N₄ [605]. To be more specific, ultrathin sulfur-doped g-C₃N₄ porous nanosheets revealed a superior photocatalytic nitrogen fixation rate with 5.99 mM·h⁻¹·g⁻¹ for 4 h under simulated sunlight irradiation [606]. Zhang et al. demonstrated that B atoms change the band structures and WO₃ enhances the photocatalytic activities. The quantum efficiency (QE) of the prepared composites is 0.71% for nitrogen fixation at 400 nm with a yield of 450.94 μmol g⁻¹ h⁻¹ under visible light [606]. In another work, Liu and coworkers mentioned that the phosphorus-doped 1 T-MoS₂ as co-catalyst decorated nitrogen-doped g-C₃N₄ nanosheets speed up the N₂ reduction rate to 689.76 μmol L⁻¹ g⁻¹·h⁻¹ in deionized water under simulated sunlight irradiation, which is 2.59, 1.65, 1.47, and 1.30 times higher than that of pure g-C₃N₄, 1 T-MoS₂@g-C₃N₄, 1 T-MoS₂@N doped g-C₃N₄, and P doped 1 T-MoS₂@g-C₃N₄, respectively. It is noteworthy to note that MoS₂ mainly includes semiconductive 2H phase and metal octahedral 1 T phase with promising conductivity and suitable photocatalysis since this phase provides a large number of active

sites on the base and edge [607]. Researchers also used metal oxides with g-C₃N₄-based composites to improve nitrogen fixation productivity. TiO₂/SrTiO₃/g-C₃N₄ ternary heterojunction nanofibers demonstrated N₂ fixation value under-stimulated sunlight irradiation is 2192 μmol g⁻¹ h⁻¹ L⁻¹, which is 1.9 and 3.3 times better than those of TiO₂/g-C₃N₄ nanofibers and SrTiO₃/g-C₃N₄ nanofibers, respectively [605,608]. Oxidized g-C₃N₄ can also enhance the chemo selective and unselective oxidative processes, utilized in organic reactions to provide synthons required in several value-added preparations [609].

It is interesting to mention that g-C₃N₄-metal oxide-based heterojunction can be widely used to detect of different biological materials, gasses, heavy metals, organic and inorganic materials [547,610]. g-C₃N₄-metal oxide-based composites can also be widely used for other applications, especially supercapacitors and desulfurization [107,611–613].

7. Conclusions, Limitations and Challenges

To fix inadequate light-harvesting of the bare g-C₃N₄, many researchers used doping elements to improve the efficiency, while combining g-C₃N₄ with other materials, such as metal oxides are another approach to deal with this issue. The investigation of g-C₃N₄-metal oxide-based heterojunctions with engineered bandgaps, and modified surface to enhance the absorption spectrum towards the visible-light region, reduction of the charge carrier recombination, and increasing the surface adsorption and reaction are among the main scopes of investigations of g-C₃N₄ based materials. Among all types of heterojunctions mentioned in the review paper, almost all g-C₃N₄-metal oxide-based nanocomposites have a Type II or a Z-scheme system, which have been proven to be outstanding for different applications. The applications highlighted included water splitting, CO₂ reduction, photodegradation of organic pollutants, nitrogen fixation, catalysis, sensing, bacterial disinfection, energy storage, etc.

Several types of carbon-based nanomaterials such as carbon-based quantum dots (CDs) (carbon quantum dots (CQDs) and graphene quantum dots (GQDs)), which are discussed in our previous work, can be used as sensors [214]. One kind of carbon-based nanomaterials is g-C₃N₄, which is widely used to detect different cells in human biological fluids, gasses, humidity, heavy metals, etc. Nevertheless, these applications also can be enormously challenging and give plenty of room for research and development, and there are great demands for fast and accurate diagnostic methods for both clinical and commercial applications. As a result, it is suggested to develop systems for clinical use to detect different diseases outside the body, especially the newly emerging one, COVID-19, mainly due to its high chemical stability and excellent efficiency in absorbing the light and promising photocatalytic performance. In addition, the detection of different heavy metals and ions is one of the most crucial factors for contaminant removal and water treatment. Since g-C₃N₄-metal oxide photocatalysts have exhibited the superior potential to detect different materials, it seems critical to extending investigations on the efficiency of these heterojunctions for detection of Cu²⁺, Hg²⁺, Cr (VI), and Ag⁺.

The need for using solar energy has increased among many researchers, as energy resources are going to be depleted. Researchers require suitable semiconductors for different catalytic reactions, such as H₂ and O₂ generation by water splitting, CO₂ reduction to hydrocarbon fuels, etc. In order to have a future practical application in this area, we have to resolve some issues about the conversion of solar energy to fuel. The cocatalysis and quantum efficiency are among the most important properties for H₂O splitting and CO₂ reduction, which for the g-C₃N₄-metal oxide composites, researchers do not have enough knowledge. The enhanced photocatalytic activity of the mentioned nanocomposites in this review under visible light encourages researchers to use these heterojunctions in antibacterial and antiviral applications. Inactivation originates from direct hole oxidation and •O₂⁻, •OH, and H₂O₂, which are produced in a reductive way, and dependent on the type of microorganisms. Because several research works have been conducted on water disinfection and microbial control in the laboratories, it is necessary to have more practical exploration on full-scale water treatment.

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