

REVIEW

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# g-C<sub>3</sub>N<sub>4</sub>-based photocatalysts for organic pollutant removal: a critical review

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## Abstract

With rapid global industrialization, environmental pollution has become one of the major problems of human society. Photocatalysis can be applied to solve environmental problem by using inexhaustible solar energy. As a novel two-dimensional material, graphite carbon nitride (g-C<sub>3</sub>N<sub>4</sub>) has been widely applied in many photocatalytic reactions. However, in terms of its practical applications, g-C<sub>3</sub>N<sub>4</sub> still has limitations, including poor photoexcited charge separation, limited range of visible light absorption, and low surface areas. Hence, current research on g-C<sub>3</sub>N<sub>4</sub> mainly focuses on improving its photocatalytic performance. In this review, we summarize various methods to improve g-C<sub>3</sub>N<sub>4</sub>'s light absorption and photo-induced charge transfer, including vacancy engineering, morphology control, heteroatom doping, and forming heterostructure. The applications of g-C<sub>3</sub>N<sub>4</sub> and its derivatives in photodegradation of organic pollutants are also discussed in detail. Finally, the challenges and future research directions of g-C<sub>3</sub>N<sub>4</sub> photocatalysts are summarized to promote their environmental applications.

## Highlights

- Structural and optical properties of g-C<sub>3</sub>N<sub>4</sub> are summarized in detail.
- Methods for improving the photocatalytic activity of g-C<sub>3</sub>N<sub>4</sub> are reviewed.
- Applications of g-C<sub>3</sub>N<sub>4</sub> in organic pollutant removal are discussed in detail.
- Challenges and future direction of g-C<sub>3</sub>N<sub>4</sub> for degrading organic pollutants are summarized.

**Keywords** Heterostructure, Graphitic carbon nitride, Photocatalysis, Photodegradation, Water treatment

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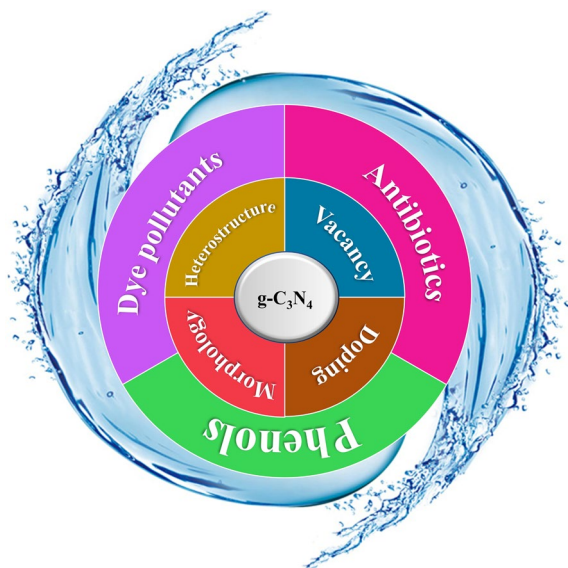
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## Graphical Abstract



## 1 Introduction

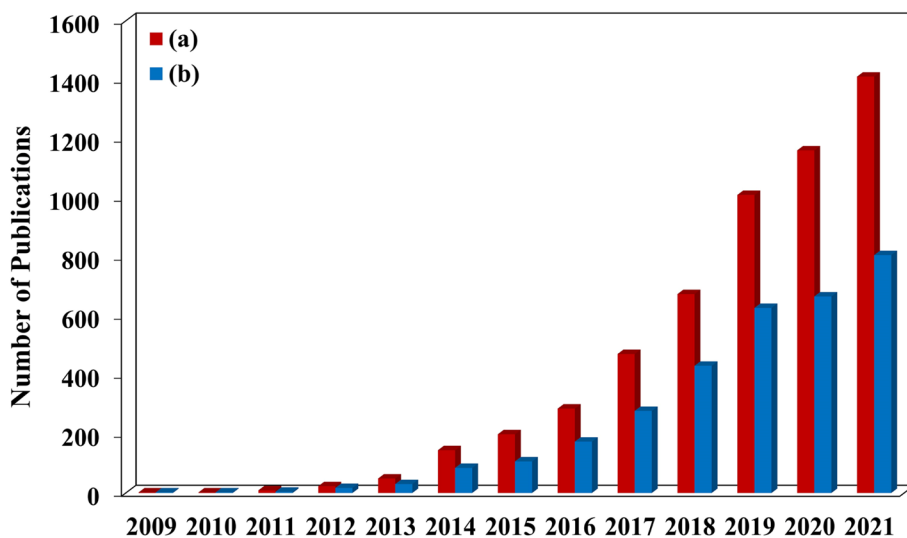
In recent decades, numerous wastewater pollutants such as dyes, phenols, and antibiotics have inevitably been released into the soil, water, rivers, and lakes, which have a negative effect on human health and ecosystem (Qiu et al. 2021). Many water treatment technologies including precipitation, biodegradation, filtration, advanced oxidation, adsorption, and photocatalysis are widely used to remove pollutants from water (Lu et al. 2022b). Among them, photocatalysis has significant applications in water purification due to its low cost, environmental friendliness, and strong removal ability.

In 1972, Fujishima and Honda found that under light irradiation, water can be decomposed into  $O_2$  and  $H_2$  in an electrochemical cell (Fujishima and Honda 1972). Since then, photocatalysis has been widely reported. Extensive research about the applications of heterostructure photocatalysis has been carried out, such as water treatment, water splitting, volatile organic compound (VOC) removal, and  $CO_2$  reduction to fuels (Liu et al. 2022; Zhang et al. 2022b). Photocatalysis happens when semiconductors absorb the energy of photons to generate electron–hole pairs, and then they quickly separate electron–hole pairs to produce active radicals for redox reactions such as degradation of organic pollutants (Lu et al. 2022a; Luo et al. 2022c). As a novel C and N based two-dimensional material, graphitic carbon nitride ( $g-C_3N_4$ ) is regarded as a new generation of

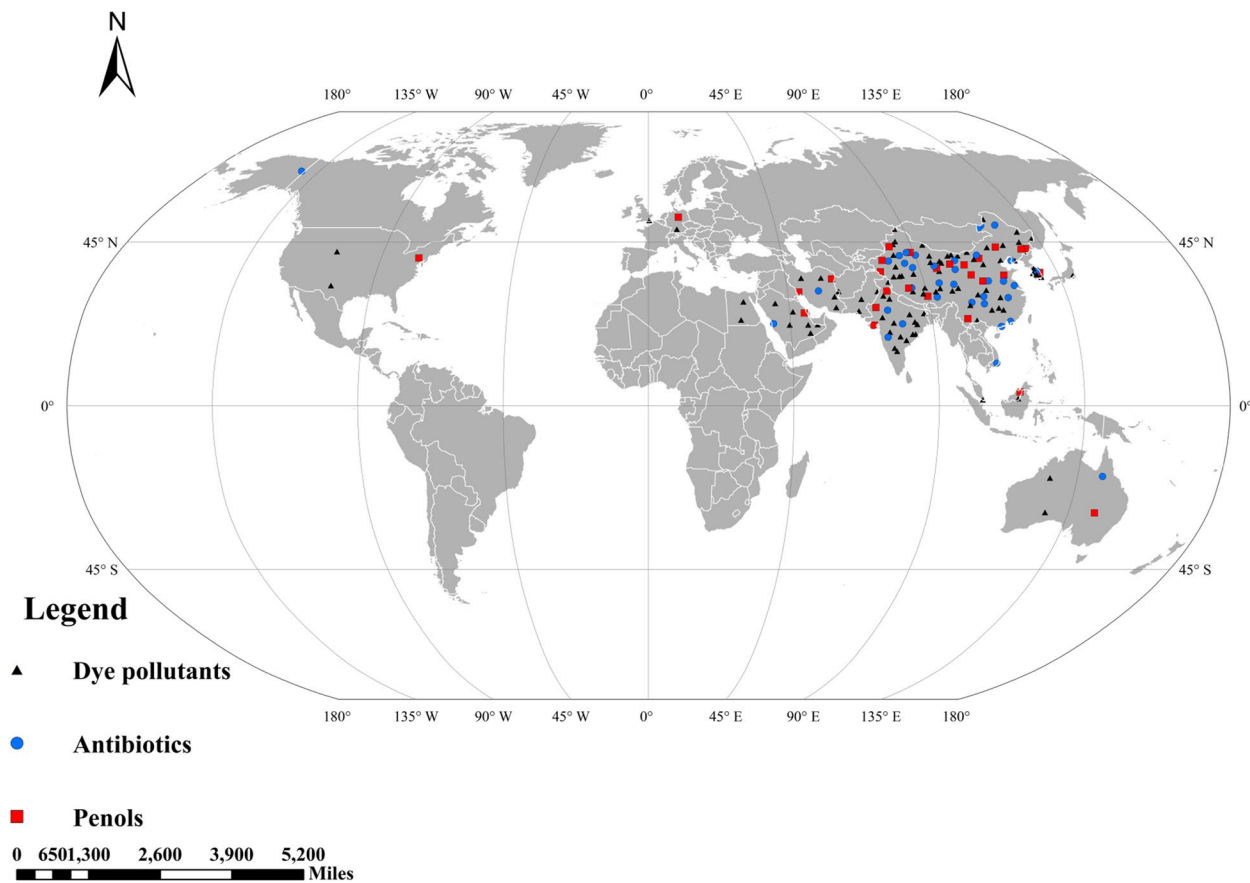
photocatalyst and has been widely used in the field of environmental photocatalysis.

Recent research trend on  $g-C_3N_4$  photocatalyst from the “Web of Science” database is displayed in Fig. 1. The search was conducted using the keywords “ $g-C_3N_4$ ” and “photocatalysis”, which generated a total of 5462 articles in 2009–2021. When “water” was added as another keyword, the number of searched articles was 3130. The results were classified by year. We can see that  $g-C_3N_4$  has been receiving increased attention for water treatment in the past decade. Fewer than five articles were published before 2011, and then the number increased significantly from 2012, and reached 771 articles in 2021.

Figure 2 shows the global distribution of papers about the  $g-C_3N_4$ -based systems for the removal of dyes, antibiotics, and phenols. The number of papers from different countries and their spatial distribution can be visualized. The number of papers is revealed using point distribution, where a point equals 20 papers. It is possible that some papers were double-counted because there were cases of co-authorship by authors from different countries. These countries were mainly distributed in Asia and Europe. In Asia, countries like China, India, Iran, Pakistan, South Korea, and Japan had many papers. The papers from Europe were mainly distributed among France, Spain, and Germany. In addition, the United States and Australia also published a considerable



**Fig. 1** a Number of annual publications using “g-C<sub>3</sub>N<sub>4</sub>” and “photocatalyst” as keywords; b Number of annual publications using “g-C<sub>3</sub>N<sub>4</sub>”, “photocatalyst”, and “water” as keywords. (Obtained from the “Web of Science”)



**Fig. 2** The global distribution of the number of papers on g-C<sub>3</sub>N<sub>4</sub>-based photocatalysts for the removal of dyes, antibiotics, and phenols

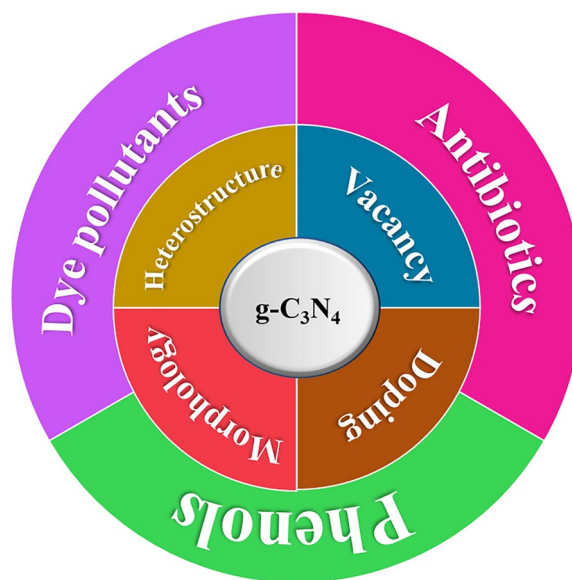
number of papers. It is easy to see that  $g\text{-C}_3\text{N}_4$ -based photocatalyst is a hot research topic worldwide.

Nitrogen-rich precursors including urea, thiourea, dicyandiamide, melamine, etc. are usually used for the synthesis of  $g\text{-C}_3\text{N}_4$  by the thermal polymerization method (Ong et al. 2016). The morphology of  $g\text{-C}_3\text{N}_4$  is similar to graphite which shows a two-dimensional layered microstructure. The tri-s-triazine structure of  $g\text{-C}_3\text{N}_4$  is the most stable phase (Zheng et al. 2015), which is regarded as the typical formation of  $g\text{-C}_3\text{N}_4$ . The  $\pi$ -conjugated structure of  $g\text{-C}_3\text{N}_4$  is  $sp^2$ -hybridized by carbon and nitrogen. The ideal C/N molar ratio of  $g\text{-C}_3\text{N}_4$  is 0.75, but most reported  $g\text{-C}_3\text{N}_4$  is rich with defects, and the C/N molar ratio is close to 0.75, which makes it more activated in catalysis. The elements C and N are earth-abundant, resulting in low-cost preparation. A small amount of hydrogen of amine groups existed in  $g\text{-C}_3\text{N}_4$ . The surface defects and hydrogen are important in photocatalysis, which can serve as surface active sites for reactants adsorption and photocatalysis (Luo et al. 2023). The bandgap of  $g\text{-C}_3\text{N}_4$  is around 2.7 eV with the conduction band (CB) and valence band (VB) potentials at ca. -1.1 eV and 1.6 eV, respectively, resulting in the visible light driven photodegradation.

However, pristine  $g\text{-C}_3\text{N}_4$  has some disadvantages, including a narrow visible-light response range, small surface areas, and a low separation rate of electron–hole pairs. All these adverse factors significantly reduce photocatalytic efficiency (Tan et al. 2021). To enhance the applications of  $g\text{-C}_3\text{N}_4$  in photocatalysis, various ways have been developed to improve its abilities for light adsorption and transfer of electron–hole pairs, including morphology control, the introduction of defects, doping with other atoms, coupling with metal, semiconductor, and carbonaceous materials (Fig. 3). To date, there are many photocatalytic applications of  $g\text{-C}_3\text{N}_4$  in wastewater treatment, including removal of dyes, antibiotics, phenols, etc. This review mainly focuses on: (1) summarizing recent development of design and preparation of  $g\text{-C}_3\text{N}_4$ -based photocatalysts, (2) systematically reviewing the photocatalytic mechanisms on different water pollutants, (3) outlining future challenges and prospects of  $g\text{-C}_3\text{N}_4$ -based photocatalysts for pollutant degradation.

## 2 $g\text{-C}_3\text{N}_4$ -based photocatalysts

Many methods have been developed to prepare  $g\text{-C}_3\text{N}_4$ . Among them, the thermal polymerization method is the most common method. In general,  $g\text{-C}_3\text{N}_4$  is easily synthesized by thermal polymerization of nitrogen-rich precursors including urea (Li et al. 2021b), melamine (Xu et al. 2020a), dicyandiamide (Bai et al. 2014), cyanamide (Liu et al. 2014b), and thiourea (Dong et al. 2013) at different temperatures. Figure 4 shows the temperature

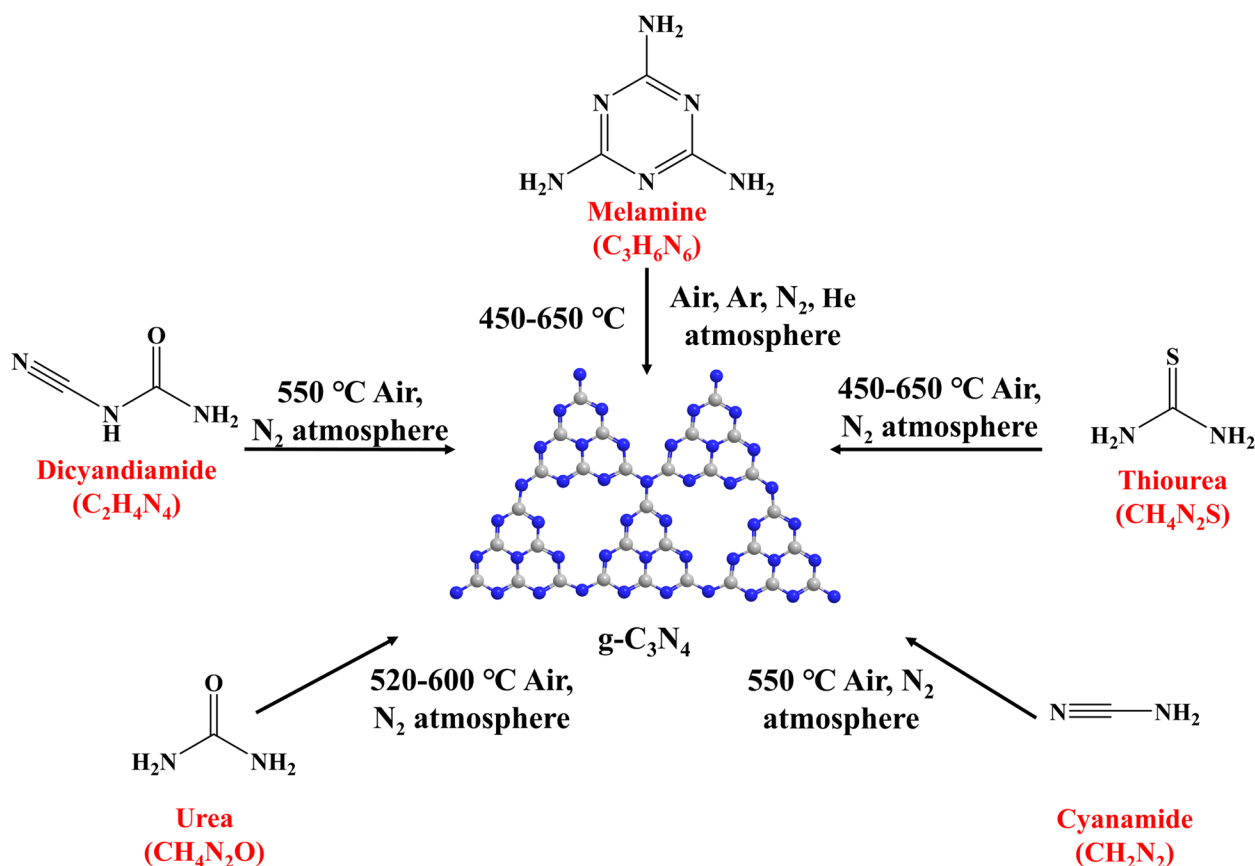


**Fig. 3** Modification methods and applications of  $g\text{-C}_3\text{N}_4$ -based photocatalysts

and atmosphere for preparing  $g\text{-C}_3\text{N}_4$  from different precursors.

### 2.1 Vacancy engineering

To improve the photocatalytic efficiency of  $g\text{-C}_3\text{N}_4$ , vacancy engineering is an effective way.  $g\text{-C}_3\text{N}_4$  with nitrogen deficiency was successfully prepared by thermal treatment of the mixture of Melamine and NaOH (Luo et al. 2021). After heating with NaOH, the N/C atomic ratio was slightly reduced according to the organic elemental analyzer and X-ray photoelectron spectroscopy (XPS) results. The existence of nitrogen vacancies in  $g\text{-C}_3\text{N}_4$  served as centers for oxidation, reduction, and charge recombination, which promoted the generation of  $h^+$ ,  $\bullet\text{O}_2^-$ , and  $^1\text{O}_2$ , thus generating  $\text{H}_2\text{O}_2$  in five different ways. According to the research of Wang et al. (2021a), N vacancy modification on  $g\text{-C}_3\text{N}_4$  nanotube can promote the activation of peroxymonosulfate (PMS) with visible-light irradiation, and perform well in real water background and a wide pH range. N vacancy in  $g\text{-C}_3\text{N}_4$  nanotube enhanced PMS adsorption and reduced it into  $\bullet\text{OH}$ , which was the major oxidant. The C atoms near N vacancy showed electron-deficient and promoted the generation of  $\bullet\text{O}_2^-$ . Liang et al. (2018) reported C vacancies in  $g\text{-C}_3\text{N}_4$  promoted the photocatalytic removal ability of Bisphenol A (BPA). C vacancies in  $g\text{-C}_3\text{N}_4$  trapped photoinduced electrons and diminished the recombination of photoinduced carriers. The trapped photo-generated electrons in carbon vacancies transferred to  $\text{O}_2$  to form  $\bullet\text{O}_2^-$ , which improved the BPA removal efficiency. According to Gao's research (2021), the CB of  $g\text{-C}_3\text{N}_4$



**Fig. 4** Preparation of g-C<sub>3</sub>N<sub>4</sub> from different precursors (Sudhaik et al. 2018; Abu-Sari et al. 2022; Nguyen et al. 2022)

moved to higher energy due to the C vacancy. The C vacancy modified g-C<sub>3</sub>N<sub>4</sub> showed 57 folds of hydrogen production efficiency and higher degradation efficiency for many pollutants such as rhodamine B (RhB), tetracycline (TC), norfloxacin (NOR), ciprofloxacin (CPX), and levofloxacin.

Based on the above results, we can conclude that C and N vacancies in g-C<sub>3</sub>N<sub>4</sub> may serve as active sites for the reactant's adsorption, promote the separation of photo-induced carriers, and change the band structure, which enhance the photocatalytic removal ability of many pollutants.

## 2.2 Morphology control

Morphology engineering of g-C<sub>3</sub>N<sub>4</sub> is a vital area of current research. Many interesting morphologies of g-C<sub>3</sub>N<sub>4</sub> have been successfully synthesized, including nanosphere, nanotube, nanosheet, and nanoflower. The g-C<sub>3</sub>N<sub>4</sub> hollow nanospheres decorated with Pt were prepared by the thermal polymerization method with a silica template and cyanamide as the precursor (Benisti et al. 2021). In the O<sub>2</sub> generation, the maximal apparent quantum efficiency of g-C<sub>3</sub>N<sub>4</sub> nanospheres was 12–15% and

improved to 40–45% after loading with Pt. The g-C<sub>3</sub>N<sub>4</sub> nanospheres were steady during 40 h of illumination. Liu et al. (2017) synthesized g-C<sub>3</sub>N<sub>4</sub> nanorods with porous shells by thermal condensation of cyanamide in silica nanotube template and further etched by HF solution. The nanorods showed enhanced photocatalytic activity and good stability in water splitting and degradation of RhB. g-C<sub>3</sub>N<sub>4</sub> can absorb the light with a wavelength of less than 475 nm (Li et al. 2020c). The bandgap will increase with the reduction of the number of nanosheets, resulting in a quantum confinement effect. Many methods were reported to diminish the thickness of the g-C<sub>3</sub>N<sub>4</sub> sheets to a few atomic layers. Thermal oxidation can etch bulk g-C<sub>3</sub>N<sub>4</sub> to 2 nm in thickness, according to the research of Niu et al. (Niu et al. 2012). The obtained anisotropic two dimensions (2D) g-C<sub>3</sub>N<sub>4</sub> nanosheets have a higher specific surface area (306 m<sup>2</sup>/g), larger bandgap, enhanced in-plane electron transfer ability, and increased photo-induced carriers' lifetime. Acid or alkali stripping is also an effective method to separate the g-C<sub>3</sub>N<sub>4</sub> layers into thin nanosheets (Taizo et al. 2013; Cui et al. 2018). The g-C<sub>3</sub>N<sub>4</sub> exfoliated by H<sub>2</sub>SO<sub>4</sub> and HNO<sub>3</sub> extended the bandgap and drastically prolonged the recombination

of photo-induced carriers (Leong et al. 2018), and it showed improved photoactivity in the removal of BPA under direct sunlight. Compared with bulk  $g\text{-C}_3\text{N}_4$ , the alkali-etched  $g\text{-C}_3\text{N}_4$  exhibited thinner and more porous  $g\text{-C}_3\text{N}_4$  nanosheets with larger specific surface area (Feng et al. 2017a), more exposed active sites, shorter carrier diffusion length, faster efficiencies of RhB degradation and hydrogen production. Mechanical strategies including ultrasonic stripping and ball milling were applied to decrease the particle size and stacked layers in  $g\text{-C}_3\text{N}_4$  nanosheets. The photodegradation of RhB on atomic single layer  $g\text{-C}_3\text{N}_4$  prepared by the ultrasonic exfoliating process was 10.2 times and 3.0 higher than that on the  $g\text{-C}_3\text{N}_4$  nanosheets with bulk morphology and few layers (Zhao et al. 2013). Ultrathin and easily dispersed  $g\text{-C}_3\text{N}_4$  nanosheets prepared by wet ball milling exhibited 2.2 times photocatalytic activity of RhB compared to untreated  $g\text{-C}_3\text{N}_4$  nanosheets (Ma et al. 2021b).

The morphology engineering of  $g\text{-C}_3\text{N}_4$  is an effective way to obtain specific properties. For example, one-dimensional (1D) rod structure can effectively improve the separation efficiency of photogenerated carriers. Two-dimensional (2D) structure usually creates large specific surface area for more active adsorption sites for photocatalytic degradation. Three-dimensional (3D) structure makes  $g\text{-C}_3\text{N}_4$  have great application potential in the field of photodegradation due to the advantages of stable photocatalytic performance and easy-recycling ability. The information is expected to provide insights into the subsequent design of  $g\text{-C}_3\text{N}_4$ -based systems with specific properties.

### 2.3 Heteroatom doping

Element doping can not only adjust the energy band structure of  $g\text{-C}_3\text{N}_4$  by adjusting the VB and CB, but also affect the surface properties of the photocatalyst, thereby improving its photocatalytic performance (Xing et al. 2022). By introducing non-metallic elements with different electronegativity and atomic radius from C and N elements, the charge redistribution of  $g\text{-C}_3\text{N}_4$  will be changed, which may lead to a change of the photocatalytic effect. Moreover, the introduction of non-metallic elements can increase the delocalization of  $\pi$ -conjugated electrons, resulting in a decrease in the band gap to improve the utilization of visible light (Tang et al. 2023). The introduction of metal impurities can produce additional binding effects and form a metal-Nx active site, enhancing the charge transfer capacity of the photocatalyst, reducing its band gap, and enhancing its adsorption capacity, thereby improving the photocatalytic activity (Xing et al. 2022).

Metal or non-metal atoms doped  $g\text{-C}_3\text{N}_4$  can be easily prepared to improve their light absorbance and

tune band energies for photocatalytic reactions. Doping many non-metallic elements like B, O, F, P, S, Cl, Br, and I was reported as an effective method to promote photocatalytic efficiency (Luo et al. 2016; Feng et al. 2019; Liu et al. 2019; Mian et al. 2020; Hu et al. 2020). The O doped  $g\text{-C}_3\text{N}_4$  was obtained with double oxidation of the concentrated acid-ultrasound method (Yang and Bian 2021). The sufficient oxygen-containing functional groups and pit-like defects were formed on the surface of  $g\text{-C}_3\text{N}_4$  by an appropriate degree of oxidation, thereby enhancing photocatalytic performance. The oxygen doping changed the main active species of  $g\text{-C}_3\text{N}_4$  in the photodegradation of RhB from  $h^+$  to  $^1\text{O}_2$ . The photocatalytic degradation efficiency of C and O doped  $g\text{-C}_3\text{N}_4$  for indomethacin was 5.9 times higher than bulk  $g\text{-C}_3\text{N}_4$ , and the main reactive species were  $^1\text{O}_2$  and  $\cdot\text{O}_2^-$  (Zheng et al. 2020). The P, O co-doped  $g\text{-C}_3\text{N}_4$  was successfully synthesized by a facile thermal polymerization method, and its degradation rate for enrofloxacin was 6.2 times higher than that of  $g\text{-C}_3\text{N}_4$  (Huang et al. 2019). Based on the nuclear magnetic resonance spectroscopy and XPS, some carbon sites were replaced by P atoms, and the nitrogen sites in the framework of  $g\text{-C}_3\text{N}_4$  were replaced by O atoms. The improved photocatalytic degradation of enrofloxacin was attributed to the enhanced surface area, narrow bandgap, and effective charge separation, and  $\cdot\text{O}_2^-$  was the main active species. The S and Cl co-doped  $g\text{-C}_3\text{N}_4$  nanosheets have a narrower band gap and better carriers' separation ability (Yi et al. 2020), which may be ascribed to the doping level of Cl 3p orbital according to the first-principles calculations. The S and Cl co-doping positively shifted the VB potential of CN and improved the photocatalytic activities for the degradation of 4-nitrophenol (4-NP) and RhB. In the P, S, and O-co-doped  $g\text{-C}_3\text{N}_4$  hydrogel, the C and N atoms were replaced and C-S, C-O, and P-N bonds were formed (Chu et al. 2020). The P, S, and O doping facilitated charge separation across the heptazine rings and attracted photoexcited electrons, which exhibited enhanced photocatalytic activity for methylene blue (MB) removal.

Doping with metal atoms has been widely investigated to enhance the photoactivity of  $g\text{-C}_3\text{N}_4$ . Er-doped  $g\text{-C}_3\text{N}_4$  were synthesized by thermal condensation of  $\text{Er}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$  and melamine for photodegradation of tylosin, RhB, and TC (Li et al. 2020a). The Er doping narrowed the bandgap and increased the photocatalytic activity. The leaching toxicity of Er-doped  $g\text{-C}_3\text{N}_4$  could be ignored. The Ce-doped mesoporous  $g\text{-C}_3\text{N}_4$  photocatalyst was constructed by plant growing guide and calcination method (Zhu et al. 2020). The improved degradation of 2-mercaptobenzothiazole was ascribed to the biomass

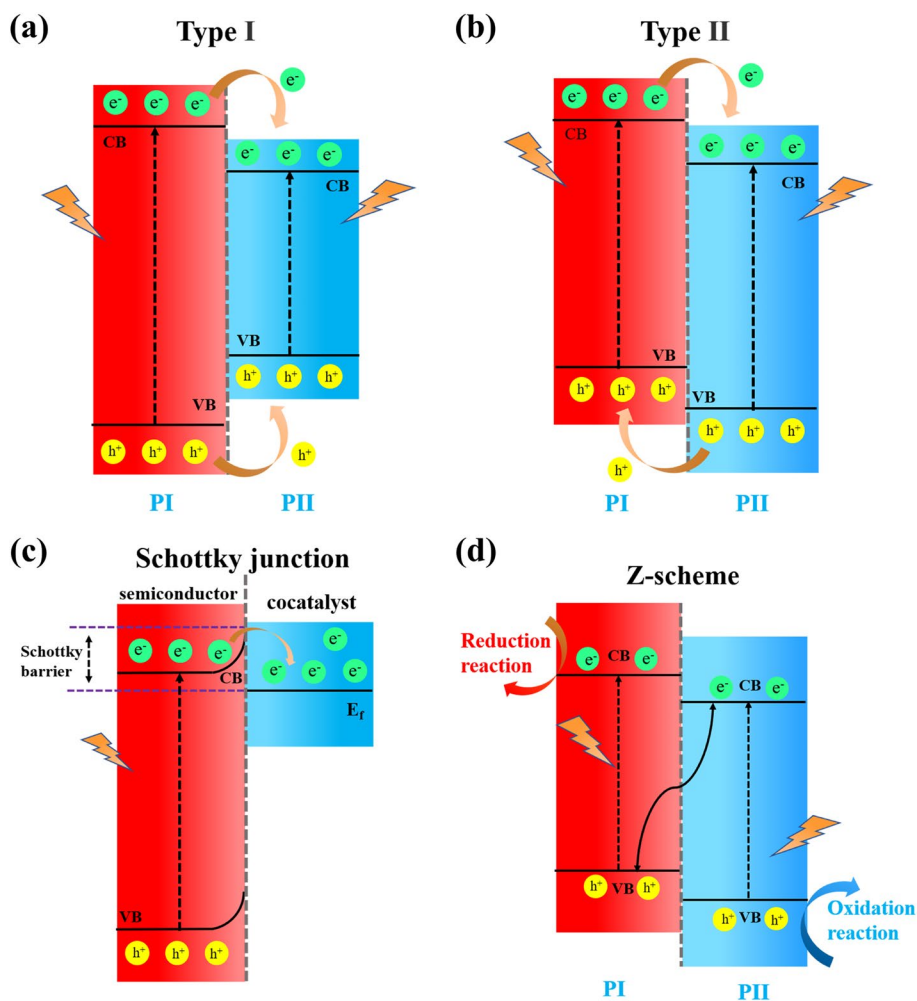
carbon enhancing the mass transport channels of carriers. The doped  $Ce^{4+}$  ions served as the redox center can degrade pollutants with photo-induced carriers.

Doping metal/nonmetal atoms to improve the performance of  $g-C_3N_4$  is a simple way to improve its photocatalytic performance. This strategy can reduce the band gap of the semiconductor to improve its optical properties and charge transfer performance. Therefore, photocatalytic abilities of metal/nonmetal atom-doped  $g-C_3N_4$  are improved and their applications in wastewater treatment are broadened.

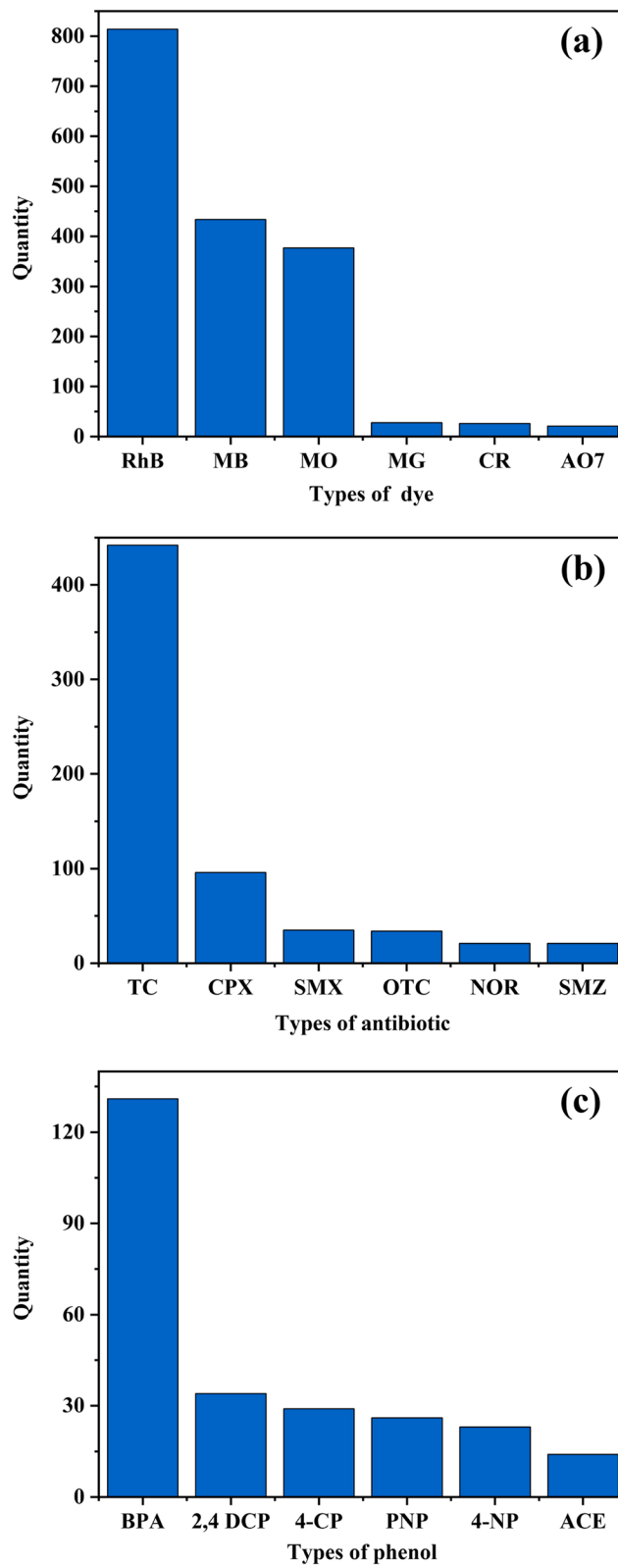
## 2.4 Heterostructure

Constructing heterostructure is another effective method to increase light absorption and enhance the photogenerated charge separation and transfer. The construction of heterojunction photocatalyst should satisfy the requirement that the coupling semiconductors have an appropriate electronic band structure to form band

arrangement. As shown in Fig. 5, the heterojunctions are classified as type I, type II, Schottky junction, and Z-scheme, which provide different charge transfer processes (Hou and Zhang 2020; Luo et al. 2022b). In the Type I heterostructure (Fig. 5a), the VB and CB of photocatalyst II (PII) are located within the band gap of photocatalyst I (PI), so electrons and holes are accumulated in photocatalyst II (PII) (Kumar et al. 2022). The type II heterostructure (Fig. 5b) could transfer electrons and holes to two different photocatalysts separately with staggered VB and CB (Yuan et al. 2021b). In the Schottky junction (Fig. 5c) (Fauzi et al. 2022), driven by the Fermi level potential difference between the two materials, photoelectrons can flow from semiconductor to cocatalyst through the interface. Therefore, the formed Schottky junction can be used as an electron trap to effectively attract photoinduced electrons, resulting in effective photoinduced charge separation. Z-scheme and type II heterojunction have similar band structures, but



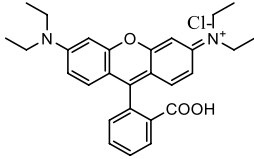
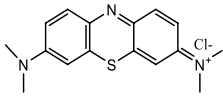
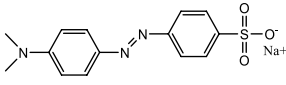
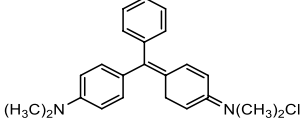
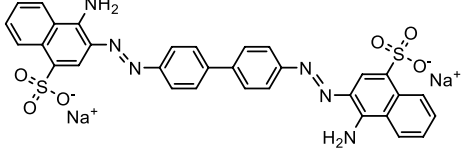
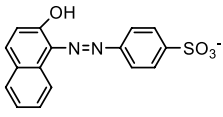
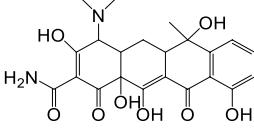
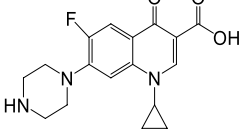
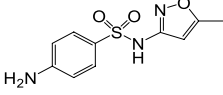
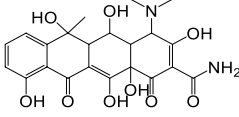
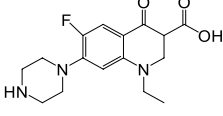
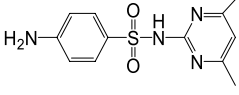
**Fig. 5** Schematic diagrams of four types of heterojunctions (Prabavathi et al. 2019; Hou and Zhang 2020; Jiang et al. 2020; Xu et al. 2020b)



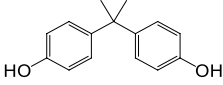
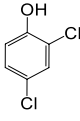
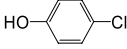
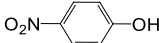
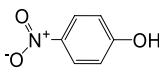
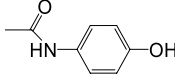
**Fig. 6** Number of papers of g-C<sub>3</sub>N<sub>4</sub>-based photocatalysts in the removal of dye pollutants (a), antibiotics (b), and phenols (c)



**Table 1** Chemical structures of various organic pollutants

Pollutants	Structure	Ref
Rhodamine (RhB)		(Jaleh et al. 2021)
Methylene blue (MB)		(Song et al. 2019)
Methyl orange (MO)		(Jaleh et al. 2021)
Malachite green (MG)		(Lin et al. 2018)
Congo red (CR)		(Jaleh et al. 2021)
Acid orange 7 (AO7)		(Wang et al. 2021d)
Tetracycline (TC)		(Zhang et al. 2021b)
Ciprofloxacin (CPX)		(Raja et al. 2020)
Sulfamethoxazole (SMX)		(Kutuzova et al. 2021)
Oxytetracycline (OTC)		(Ye et al. 2019)
Norfloxacin (NOR)		(Tian et al. 2018)
Sulfamethazine (SMZ)		(Zhang et al. 2021a)

**Table 1** (continued)

Pollutants	Structure	Ref
Bisphenol A (BPA)		(Jaleh et al. 2021)
2,4-dichlorophenol (2,4-DCP)		(Qu et al. 2020)
4-chlorophenol (4-CP)		(Jaleh et al. 2021)
P-nitrophenol (PNP)		(Darie et al. 2019)
4-nitrophenol (4-NP)		(Jaleh et al. 2021)
Acetaminophen (ACE)		(Mu et al. 2020)

the direction of electron transfer is different. As shown in Fig. 5d (Xu et al. 2018), the electrons in CB of PII will recombine with holes in VB of PI, and holes with strong oxidation ability in PII and electrons in the CB of PI with strong reduction abilities are preserved, resulting in enhanced photocatalytic activity.

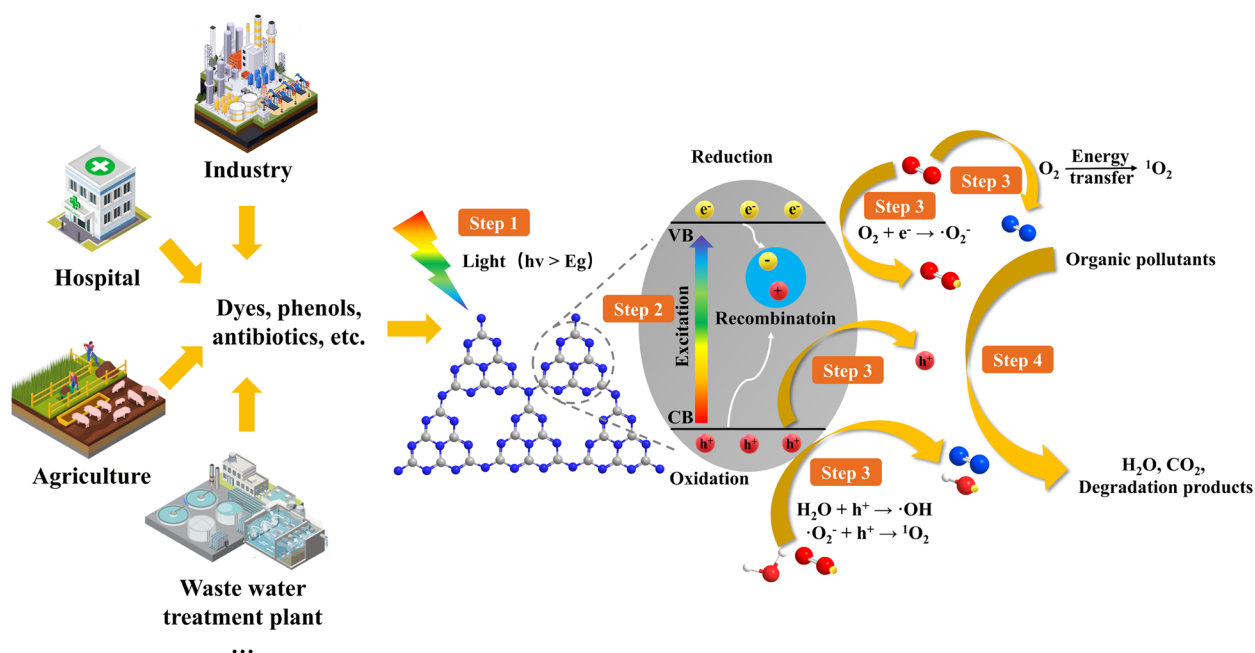
Choosing two or more semiconductor materials to construct heterojunction structure reasonably can integrate the advantages of many effects to improve the separation of photogenerated charges, expand the absorption range of visible light, and maintain the high redox ability of photocatalyst. Therefore, the design and synthesis of different types of  $g\text{-C}_3\text{N}_4$ -based heterojunction photocatalysts can effectively improve photocatalytic performance and are widely used in the field of environmental water treatment.

### 3 Photocatalytic degradation of organic pollutants

Recently, the photocatalytic applications of  $g\text{-C}_3\text{N}_4$ -based materials in the environment have attracted wide attention. In this part, the photodegradation applications of  $g\text{-C}_3\text{N}_4$ -based materials for dye pollutants, antibiotics, and phenols are reviewed. Figure 6 shows the number of publications on the removal of various dye pollutants, antibiotics, and phenols.

The structures of these pollutants are presented in Table 1. Dyes are an important pollutant, which can even be identified by naked eyes. Usually, dyes pollute the water bodies and cause skin allergy and eye irritation. Therefore, it is necessary to eliminate dye pollution in the environment. According to their chemical structure, dyes are mainly divided into azo dyes, anthraquinone

dyes, xanthene dyes, triarylmethane dyes, thiazine dyes, etc. Azo dyes such as acid orange 7 (AO7), congo red (CR), and methyl orange (MO) are cost-effective and stable (Rauf and Ashraf 2009; Raval et al. 2016). Compared with azo dyes, anthraquinone dyes such as pigment yellow 108 are less stable and more expensive (Gupta and Suhas 2009). The xanthene dyes such as RhB usually exhibit yellow, pink or red color depending on the functional group. Triarylmethane dyes such as malachite green (MG) are synthetic organic compounds with three aryl groups connected to the central carbon atom (Rauf and Ashraf 2009). Thiazine dyes such as MB are mostly used dyes for laboratory uses (Kiernan and Histochemistry 2001; Le et al. 2022). According to their chemical structures, antibiotics can be divided into  $\beta$ -lactams, aminoglycosides, macrolides, amide alcohols, tetracyclines, polypeptides, lincosamides, polyphosphates, fluoroquinolones, sulfonamides and so on (Pi et al. 2018; Biswal and Balasubramanian 2022). TC and oxytetracycline (OTC) are the most studied tetracyclines. CPX and NOR are the most concerned fluoroquinolones. In sulfonamides, sulfamethoxazole (SMX) and sulfamethazine (SMZ) have been widely studied (Margolis et al. 2010; Roca Jalil et al. 2015; Oliveira et al. 2019; Chandra et al. 2021; Shurbaji et al. 2021). Antibiotics may affect the distribution of microbial communities in water and affect humans through food chain transmission. At the same time, the prevalence of antibiotics may lead to antibiotic resistance (Qin et al. 2021). Phenolic compounds have at least one aromatic ring with one or more hydroxyl groups. Common phenols are BPA, 2,4-dichlorophenol (2,4-DCP), 4-chlorophenol (4-CP),



**Fig. 7** Degradation mechanisms of  $g\text{-C}_3\text{N}_4$  for organic pollutants of different sources (Song et al. 2017; Amanulla et al. 2018; Wei et al. 2021; Zhao et al. 2021b)

p-nitrophenol (PNP), 4-NP, acetaminophen (ACE) and so on. They are toxic and carcinogenic to humans, animals, and wildlife even at low concentrations (Liu et al. 2014a). Photocatalysis as an economically feasible way for the removal of dyes, antibiotics, and phenolic pollutants has attracted widespread attention.

The sources of antibiotics, phenols, and dyes are shown in Fig. 7. So far,  $g\text{-C}_3\text{N}_4$  has been widely used as an environmental photocatalyst, therefore its photocatalytic degradation mechanism for organic pollutants has been explored. The photocatalytic process can be divided into four stages (Fig. 7). Firstly, visible light is absorbed by  $g\text{-C}_3\text{N}_4$  (Step 1). The light absorption stage is up to the surface morphology and structure of the photocatalyst (Wen et al. 2017). When the energy of light is greater than or equal to the energy of the semiconductor band gap ( $E_g$ ), the electrons are excited from VB to CB, leaving holes in VB, thus achieving effective separation of photogenerated carriers (Step 2) (Zhang et al. 2019). The band gap of  $g\text{-C}_3\text{N}_4$  can be further adjusted by doping, introducing defects, etc. (Wen et al. 2017), thus promoting its use of visible light. At the same time, in order to promote photocatalysis, it is necessary to prevent the recombination of electrons and holes. Furthermore, photogenerated electrons and holes are separated and migrated (Step 3). Photogenerated electrons react with electron acceptors (such as oxygen) to form superoxide groups, and holes react with water to produce hydroxyl

radicals (Ge et al. 2011). Singlet oxygen ( $^1\text{O}_2$ ) can be produced by the reaction of holes with superoxide groups or reacting with oxygen by energy transfer (Zhang et al. 2009; Zhou et al. 2017). At last,  $g\text{-C}_3\text{N}_4$  can adsorb the diffused pollutants on its surface, and the active substances degrade them into the water, carbon dioxide, and other products after redox reactions (Step 4). For each target pollutant, the photocatalytic mechanism will be different. The following sections summarize the degradation mechanisms for specific pollutants.

### 3.1 Degradation of dyes

Dyes pose a serious threat to the aqueous environment owing to their non-biodegradability and chemical stability in water. More than 100,000 organic dyes with an annual production of over  $7 \times 10^5$  t are commercially available (Zhang et al. 2020b). Hence, green methods to treat dye pollutants with low-cost and high efficiency are urgently needed.  $g\text{-C}_3\text{N}_4$ -based materials have unique characteristics, such as tunable nanostructures, chemical stabilities, and rich active sites, making them particularly suitable for dye degradation (Liang et al. 2021). Table 2 lists the recent  $g\text{-C}_3\text{N}_4$ -based materials for dyes degradation in wastewater.

RhB is a water-soluble xanthene dye that irritates the eyes and skin and is deleterious if swallowed by animals and human beings (Liang et al. 2021). It is the most widely studied dye worldwide (Fig. 6a). The

**Table 2** Information on the degradation of dye pollutants by g-C<sub>3</sub>N<sub>4</sub>-based photocatalyst<sup>a</sup>

Catalyst	Process conditions	Light source	Catalyst dosage	Performance	Active species	Ref
O-doped g-C <sub>3</sub> N <sub>4</sub>	RhB: 20 mg/L, 30 mL	350 W Xe lamp	30 mg	180 min, 100%	<b><sup>1</sup>O<sub>2</sub></b> , <b>•O<sub>2</sub><sup>-</sup></b> , <b>h<sup>+</sup></b>	(Yang and Bian 2021)
Co <sub>3</sub> O <sub>4</sub> @g-C <sub>3</sub> N <sub>4</sub>	RhB: 20 mg/L, 50 mL	300 W Xe lamp	10 mg	60 min, 95%	<b>•O<sub>2</sub><sup>-</sup></b> , <b>h<sup>+</sup></b> , <b>•OH</b>	(Rabani et al. 2021)
Ag-B doped g-C <sub>3</sub> N <sub>4</sub>	RhB: 10 mg/L, 50 mL	150 W Xe lamp	10 mg	80 min, 100%	<b>•OH</b> , <b>•O<sub>2</sub><sup>-</sup></b>	(Roselin et al. 2019)
P-K doped g-C <sub>3</sub> N <sub>4</sub>	RhB: 20 mg/L, 100 mL	300 W Xe lamp (λ ≥ 420 nm)	20 mg	80 min, 98.2%	<b>h<sup>+</sup></b> , <b>•O<sub>2</sub><sup>-</sup></b> , <b>•OH</b>	(Shen et al. 2020)
g-C <sub>3</sub> N <sub>4</sub> /carbon nano-tubes	RhB: 10 mg/L, 100 mL	300 W Xe lamp (λ > 420 nm)	20 mg	60 min, 98.1%	<b>•O<sub>2</sub><sup>-</sup></b> , <b>e<sup>-</sup></b> , <b>•OH</b>	(Liu et al. 2020)
BaTiO <sub>3</sub> /Au/g-C <sub>3</sub> N <sub>4</sub>	RhB: 5 mg/L, 100 mL	300 W Xe lamp	20 mg	20 min, 100%	<b>h<sup>+</sup></b> , <b>•OH</b> , <b>•O<sub>2</sub><sup>-</sup></b>	(Wu et al. 2020)
BiOBr/g-C <sub>3</sub> N <sub>4</sub>	RhB: 10 mg/L, 100 mL	100 W Xe lamp	20 mg	30 min, 95.23%	<b>•O<sub>2</sub><sup>-</sup></b> , <b>h<sup>+</sup></b> , <b>•OH</b>	(Zhang et al. 2021c)
Carbonized poly(tannic acid)/g-C <sub>3</sub> N <sub>4</sub>	RhB: 10 mg/L, 30 mL	500 W Xe lamp (λ ≥ 420 nm)	30 mg	80 min, 97%	<b>•O<sub>2</sub><sup>-</sup></b> , <b>h<sup>+</sup></b> , <b>•OH</b>	(Yang et al. 2021)
Perylene diimide-g-C <sub>3</sub> N <sub>4</sub> /MIL	RhB: 100 mg/L, 30 mL	Xe lamp	3 mg	60 min, 98%	<b>•O<sub>2</sub><sup>-</sup></b> , <b>h<sup>+</sup></b> , <b>•OH</b>	(Wang et al. 2021c)
Cd <sub>0.5</sub> Zn <sub>0.5</sub> S/g-C <sub>3</sub> N <sub>4</sub>	RhB: 4.79 mg/L, 50 mL	350 W Xe lamp (λ > 420 nm)	50 mg	90 min	<b>•O<sub>2</sub><sup>-</sup></b> , <b>h<sup>+</sup></b> , <b>•OH</b>	(Qin et al. 2020)
Bi <sub>2</sub> MoO <sub>6</sub> /g-C <sub>3</sub> N <sub>4</sub>	RhB: 5 mg/L, 100 mL	300 W Xe lamp (λ > 420 nm)	5 mg	40 min, 95.4%	<b>h<sup>+</sup></b> , <b>•O<sub>2</sub><sup>-</sup></b> , <b>•OH</b>	(Li et al. 2020b)
Li doped g-C <sub>3</sub> N <sub>4</sub> /Co <sub>3</sub> O <sub>4</sub>	RhB: 10 mg/L, 50 mL	300 W Xe lamp	50 mg	60 min, 98%	<b>h<sup>+</sup></b> , <b>•O<sub>2</sub><sup>-</sup></b> , <b>•OH</b>	(Niu et al. 2022)
g-C <sub>3</sub> N <sub>4</sub> /BiOI	RhB: 20 mg/L, 50 mL	500 W Xe lamp (λ > 420 nm)	20 mg	120 min, 99%	<b>h<sup>+</sup></b> , <b>•O<sub>2</sub><sup>-</sup></b> , <b>•OH</b>	(Hou et al. 2021)
g-C <sub>3</sub> N <sub>4</sub> /Fe <sub>3</sub> O <sub>4</sub> /RGO	RhB: 15 mg/L, 50 mL	400 W metal halide lamp (λ > 400 nm)	10 mg	75 min, 100%	<b>•O<sub>2</sub><sup>-</sup></b> , <b>•OH</b> , <b>h<sup>+</sup></b>	(Luo et al. 2022a)
N-doped carbon quantum dots (CQDs)/g-C <sub>3</sub> N <sub>4</sub>	MB: 10 mg/L, 100 mL	18 W LED lamp	50 mg	180 min, 54.6%	<b>h<sup>+</sup></b> , <b>•OH</b> , <b>•O<sub>2</sub><sup>-</sup></b>	(Seng et al. 2020)
g-C <sub>3</sub> N <sub>4</sub> /ZnO, Zr/g-C <sub>3</sub> N <sub>4</sub>	MB: 10 mg/L, 200 mL	350 W Xe lamp (400 ≤ λ < 800 nm)	100 mg	180 min, 98.07%	<b>h<sup>+</sup></b> , <b>•O<sub>2</sub><sup>-</sup></b> , <b>•OH</b>	(Guo et al. 2020)
Ni-ZnS/g-C <sub>3</sub> N <sub>4</sub>	MB: 10 mg/L, 220 mL	500 W tungsten halogen direct lamp (λ ≥ 400 nm)	220 mg	40 min, 96%	<b>•O<sub>2</sub><sup>-</sup></b> , <b>•OH</b> , <b>h<sup>+</sup></b>	(Danish and Muneer 2021)
S-doped g-C <sub>3</sub> N <sub>4</sub> /TiO <sub>2</sub>	CR: 50 mg/L, 100 mL	300 W Xe lamp	20 mg	60 min, 96.2%	<b>•O<sub>2</sub><sup>-</sup></b> , <b>•OH</b> , <b>h<sup>+</sup></b>	(Wang et al. 2021b)
g-C <sub>3</sub> N <sub>4</sub> /ZnSe	CR: 100 mg/L, 80 mL	300 W Xe lamp (λ > 420 nm)	10 mg	60 min, 95.69%	<b>•O<sub>2</sub><sup>-</sup></b> , <b>•OH</b> , <b><sup>1</sup>O<sub>2</sub></b>	(Ehsan et al. 2020)
BiOI-BiOCl/g-C <sub>3</sub> N <sub>4</sub>	AO7: 10 mg/L, 100 mL	400 W halogen lamp	100 mg	140 min, 97.4%	<b>h<sup>+</sup></b> , <b>•O<sub>2</sub><sup>-</sup></b> , <b>•OH</b>	(Aghdam et al. 2017)

<sup>a</sup> The main active species in each reference are shown boldly in the table

SiO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> (Si-CN) composite photocatalyst and surface hydroxylation modified SiO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> (Si-CN-HO) were prepared for RhB degradation (Sun et al. 2021a, b). Firstly, SiO<sub>2</sub> and melamine are ground separately with ethanol to obtain two mixed slurries. Secondly, the two mixed slurries were grounded together and calcined in a muffle furnace to prepare Si-CN composite photocatalyst. The prepared Si-CN composites were refluxed in H<sub>2</sub>O<sub>2</sub> solution to realize the surface hydroxylation modification of Si-CN-HO. The photocatalytic efficiency of RhB by hydroxylated Si-CN was 20.9 times of that by CN under solar light irradiation. Scanning electron microscopy (SEM) and transmission electron microscope (TEM) analyses showed that combining

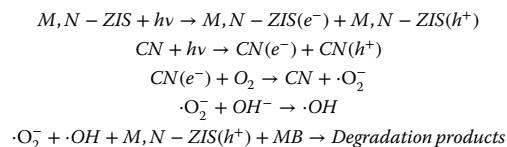
SiO<sub>2</sub> and CN significantly suppressed the clustering of CN nanosheets, in which SiO<sub>2</sub> acted as a space barrier to CN. In addition, hydroxyl-modified Si-CN further decreased the agglomeration of CN nanosheets, which could be ascribed to the escape of oxygen during surface modification. The specific surface area of Si-CN was 1.8 times of that of CN. It could be concluded from UV–Vis absorption spectra that Si-CN had higher light absorption intensity, due to the improvement of dispersion. The optical absorption intensity of Si-CN-HO was even better than that of Si-CN, which increased with the modification temperature. This might be due to the strong oxidation of H<sub>2</sub>O<sub>2</sub> that caused some defects on the surface of CN, thus forming impurity levels and

improving the optical absorption capacity. In addition, the capture experiments confirmed that  $\bullet\text{O}_2^-$  is the main active group for the photocatalytic degradation of RhB. The reaction mechanism reveals the reason for the photocatalytic activity enhancement of Si-CN-HO samples. The electrons on the VB of CN were excited by light and transited to the CB in Si-CN-HO. The holes left in the VB migrated to the semiconductor surface and were trapped by hydroxyl groups. At the same time, the electrons were trapped by oxygen to form  $\bullet\text{O}_2^-$ , which further degraded RhB.

The surface hydroxylation treatment of Si-CN composite led to the dispersion of  $\text{SiO}_2$  on CN by enhancing the combination of  $\text{SiO}_2$  and CN, resulting in an obvious improvement in surface area. This facilitated the adsorption of pollutants by Si-CN-HO, thus promoting pollutant degradation. In addition, the optical absorption performance of Si-CN-HO was better than that of bulk CN due to the optical reflection effect from  $\text{SiO}_2$  and surface defects created by surface hydroxylation treatment, which improved the optical utilization efficiency in photocatalytic reaction.

The formation of heterojunction is an effective strategy to improve photocatalytic activity. Mo, N co-doped  $\text{ZnIn}_2\text{S}_4/\text{g-C}_3\text{N}_4$  material (M, N-ZIS/CN) was prepared by sol-gel method for photodegradation of MB (Ma et al. 2022). Firstly,  $\text{g-C}_3\text{N}_4$  was obtained by calcination using a three-step procedure. Then, zinc acetate and indium chloride were added to 150 mL distilled water and stirred for 30 min before thioacetamide was added. The  $\text{g-C}_3\text{N}_4$  dispersed in distilled water was then added drop by drop. The solution was heated and dried to form a gel. The dried gel was washed and vacuum dried to obtain  $\text{ZnIn}_2\text{S}_4/\text{g-C}_3\text{N}_4$  material. M, N-ZIS/CN nanosheets were obtained by adding ammonium molybdate and N-N-dimethylformamide to the solution for the preparation of  $\text{ZnIn}_2\text{S}_4/\text{g-C}_3\text{N}_4$ , and the rest procedures were the same as the preparation method of  $\text{ZnIn}_2\text{S}_4/\text{g-C}_3\text{N}_4$  material. SEM and TEM analyses showed that  $\text{ZnIn}_2\text{S}_4$  was a thick nanoflower structure,  $\text{g-C}_3\text{N}_4$  was thin nanosheets, and  $\text{ZnIn}_2\text{S}_4/\text{g-C}_3\text{N}_4$  was thin nanosheets, but M, N-ZIS/CN was small and loose 2D nanoflower. These indicate that co-doping increased the specific surface area of the composite material, increased its active site, shortened the charge transfer path, and reduced the agglomeration of  $\text{ZnIn}_2\text{S}_4$  particles. UV-Vis absorption spectra showed that M, N-ZIS/CN had a wide absorption range in ultraviolet and visible light. Photocatalytic tests showed that M, N-ZIS/CN could degrade 97% of MB in 120 min, which was 2.6 times that of  $\text{ZnIn}_2\text{S}_4$ . In the cycle test, M, N-ZIS/CN used for the third time could still degrade 95.2% of MB, indicating that M, N-ZIS/CN had strong photocatalytic

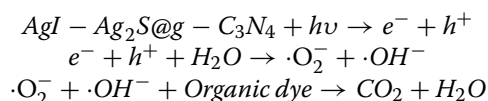
degradation ability and stability. The radicals trapping experiment found that  $e^-$ ,  $\bullet\text{O}_2^-$  and  $h^+$  were the main active species of degrading pollutants, and their degradation mechanism was as follows:



The Z-scheme heterostructure of M, N-ZIS/CN was prepared by sol-gel method. The internal electric field formed by the material accelerated the migration of charges and promoted the redox reaction.

Designing ternary  $\text{g-C}_3\text{N}_4$ -based composite heterostructure to improve photocatalytic performance is also booming. AgI-Ag<sub>2</sub>S impregnated  $\text{g-C}_3\text{N}_4$  (AgI-Ag<sub>2</sub>S@g-C<sub>3</sub>N<sub>4</sub>) composites were synthesized by hydrothermal and pyrolysis methods (Velmurugan et al. 2020).  $\text{g-C}_3\text{N}_4$  was prepared by thermal polymerization of dicyandiamide. AgI-Ag<sub>2</sub>S was prepared by wet chemical route. Finally,  $\text{g-C}_3\text{N}_4$  was ultrasonically dispersed in a methanol solution, and then a certain amount of AgI-Ag<sub>2</sub>S powder was added to the above solution and stirred and heated until the methanol was completely evaporated to obtain AgI-Ag<sub>2</sub>S@g-C<sub>3</sub>N<sub>4</sub> photocatalyst. Based on the nitrogen adsorption and desorption isotherms, the surface area of AgI-Ag<sub>2</sub>S@g-C<sub>3</sub>N<sub>4</sub> nanocomposites increased, owing to the fact that AgI-Ag<sub>2</sub>S can prevent the agglomeration of  $\text{g-C}_3\text{N}_4$ , thereby improving the adsorption of dyes on the active surface of the catalyst and enhancing the interaction between the photocatalyst and the pollutants. In addition, the optical absorption properties of the samples were analyzed by UV-Vis diffuse reflectance spectroscopy. Compared with the  $\text{g-C}_3\text{N}_4$ , the band gap of AgI-Ag<sub>2</sub>S@g-C<sub>3</sub>N<sub>4</sub> was significantly reduced and the light response range was increased, beneficial to the separation of photogenerated electron holes. The photogenerated charge separation was further studied by photoluminescence spectroscopy (PL). On the one hand, AgI-Ag<sub>2</sub>S@g-C<sub>3</sub>N<sub>4</sub> had a lower PL intensity, indicating that there was a lower photogenerated charge recombination rate in the composite material. On the other hand, with the slight shift of the characteristic band of  $\text{g-C}_3\text{N}_4$  in the Raman spectrum of AgI-Ag<sub>2</sub>S@g-C<sub>3</sub>N<sub>4</sub> nanocomposites, the signal of  $\text{g-C}_3\text{N}_4$  was highly enhanced, indicating that AgI-Ag<sub>2</sub>S had strong adsorption on the  $\text{g-C}_3\text{N}_4$  surface by charge transfer resonance. With Evans Blue (EB) and congo red as target contaminants, AgI-Ag<sub>2</sub>S@g-C<sub>3</sub>N<sub>4</sub> achieved 98.4% of EB degradation in 50 min and 94.2% of CR degradation in 30 min. In order to clarify the reasons for the enhancement of photocatalytic activity, the researchers explored its photocatalytic mechanism. It was found that a double Z-scheme heterojunction of  $\text{g-C}_3\text{N}_4$ -Ag<sub>2</sub>S

and Ag<sub>2</sub>S-AgI was formed in AgI-Ag<sub>2</sub>S@g-C<sub>3</sub>N<sub>4</sub>, which effectively inhibited the recombination of photogenerated carriers, so that more active substances were used for photocatalytic reactions. The specific photocatalytic degradation process of organic dyes on AgI-Ag<sub>2</sub>S@g-C<sub>3</sub>N<sub>4</sub> nanocomposites was as follows:



Besides, AgI-Ag<sub>2</sub>S@g-C<sub>3</sub>N<sub>4</sub> had excellent cycle stability after dye photodegradation, indicating its sustainability in future environmental purification

prospects. g-C<sub>3</sub>N<sub>4</sub>-based composite photocatalysts have been widely used in the photocatalytic degradation of various dyes in water. These composites adsorb dyes onto their surfaces and then degrade them under visible light excitation. Combining g-C<sub>3</sub>N<sub>4</sub> with other materials to form different types of heterojunction structures can effectively increase the specific surface area, provide more reduction reaction sites, and improve the light response range and charge separation.

### 3.2 Degradation of antibiotics

Antibiotics are crucial for the treatment of infectious diseases worldwide. However, a large number of antibiotics

**Table 3** Information on the degradation of antibiotics by g-C<sub>3</sub>N<sub>4</sub>-based photocatalyst<sup>a</sup>

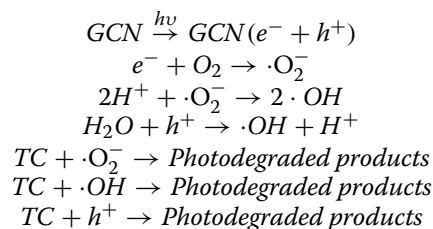
Catalyst	Process conditions	Light source	Catalyst dosage	Performance	Active species	Ref
C-doped g-C <sub>3</sub> N <sub>4</sub> /WO <sub>3</sub>	TC: 10 mg/L, 100 mL	500 W Xe lamp (λ > 420 nm)	100 mg	60 min, 75%	•O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , •OH	(Zhao et al. 2021a)
Bi <sub>2</sub> O <sub>3</sub> /CO <sub>3</sub> /S-doped g-C <sub>3</sub> N <sub>4</sub>	TC: 10 mg/L	300 W Xe lamp	30 mg	30 min, 82.6%	h <sup>+</sup> , •O <sub>2</sub> <sup>-</sup> , •OH	(Liu et al. 2021b)
CeO <sub>2</sub> /N-doped CQDs g-C <sub>3</sub> N <sub>4</sub>	TC: 20 mg/L, 100 mL	300 W Xe lamp (λ > 420 nm)	100 mg	60 min, 100%	•O <sub>2</sub> <sup>-</sup> , •OH, h <sup>+</sup> , e <sup>-</sup>	(Qi et al. 2020)
PNIPAM-Ag/Ag <sub>3</sub> PO <sub>4</sub> /g-C <sub>3</sub> N <sub>4</sub>	TC: 20 mg/L, 100 mL	250 W Xe lamp	50 mg	120 min, 88.96%	•O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , •OH	(Sun et al. 2020)
Fe-doped g-C <sub>3</sub> N <sub>4</sub>	TC: 20 mg/L, 60 mL	300 W Xe lamp	30 mg	80 min, 63.7%	•O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , •OH	(Xu et al. 2019)
g-C <sub>3</sub> N <sub>4</sub> @ZIF-8	TC: 20 mM, 50 mL	300 W Xe lamp (λ ≥ 430 nm)	10 mg	60 min, 87.6%	•O <sub>2</sub> <sup>-</sup> , <sup>1</sup> O <sub>2</sub> , h <sup>+</sup> , e <sup>-</sup> , •OH	(Yuan et al. 2021a)
Mo <sub>2</sub> C/g-C <sub>3</sub> N <sub>4</sub>	TC: 20 mg/L, 30 mL	300 W Xe lamp (λ > 420 nm)	30 mg	60 min, 91.6%	•O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , •OH	(Zhang et al. 2020a)
Co <sub>3</sub> O <sub>4</sub> /g-C <sub>3</sub> N <sub>4</sub>	TC: 15 mg/L, 100 mL	500 W Xe lamp (λ > 420 nm)	40 mg	150 min, 92.6%	•O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , •OH	(Zhao et al. 2021c)
g-C <sub>3</sub> N <sub>4</sub> /Ag/BNQDs	TC: 100 mL	300 W Xe lamp (λ > 420 nm)	50 mg	80.54%, 60 min,	•O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , •OH	(Ren et al. 2022)
g-C <sub>3</sub> N <sub>4</sub> /g-C <sub>3</sub> N <sub>4-x</sub>	TC: 20 mg/L, 100 mL	300 W Xe lamp (λ > 420 nm)	50 mg	92.7%, 60 min	•O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , •OH	(Feng et al. 2023)
Ag/g-C <sub>3</sub> N <sub>4</sub>	TC: 20 mg/L, 200 mL	300 W Xe lamp (λ ≥ 420 nm)	50 mg	100%, 75 min	•O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , •OH, H <sub>2</sub> O <sub>2</sub>	(Sun et al. 2022)
AgI/(Na,F) doped g-C <sub>3</sub> N <sub>4</sub>	TC: 35 mg/L, 100 mL	300 W Xe lamp (λ > 400 nm)	50 mg	21 min	•O <sub>2</sub> <sup>-</sup> , •OH, h <sup>+</sup>	(Zhou et al. 2021)
g-C <sub>3</sub> N <sub>4</sub> /CQDs/carbon cloth	CPX: 5 mg/L			60 min, 98%	•O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , •OH	(Wang et al. 2021e)
Ag <sub>3</sub> PO <sub>4</sub> /g-C <sub>3</sub> N <sub>4</sub>	SMX: 1 mg/L, 100 mL	300 W Xe lamp (λ > 400 nm)	5 mg	60 min, 100%	h <sup>+</sup> , •O <sub>2</sub> <sup>-</sup> , •OH, <sup>1</sup> O <sub>2</sub>	(Zhou et al. 2017)
g-C <sub>3</sub> N <sub>4</sub> /Bi <sub>4</sub> NbO <sub>8</sub> Cl	OTC: 20 mg/L, 10 mL	18 W LED light	10 mg	60 min, 87%	•O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , •OH	(Majumdar et al. 2021)
Co/g-C <sub>3</sub> N <sub>4</sub>	NOR: 6 mg/L, 30 mL	500 W Xe lamp (λ > 420 nm)	40 mg	10 min, 96.4%	•OH, SO <sub>4</sub> <sup>•-</sup>	(Zhang et al. 2021d)
CuO-ZnO/g-C <sub>3</sub> N <sub>4</sub>	amoxicillin: 60 mg/L, 250 mL, pH = 7	300 W Xe lamp	225 mg	120 min, 100%	•OH, •O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , e <sup>-</sup>	(Moradi et al. 2021)
UiO-66/NH <sub>2</sub> -MIL-125/g-C <sub>3</sub> N <sub>4</sub>	OFX: 20 mg/L	300 W Xe lamp (λ > 420 nm)	0.1 g/L	99.1%, 50 min	•O <sub>2</sub> <sup>-</sup> , •OH, h <sup>+</sup> , e <sup>-</sup>	(Sepehri-mansourie et al. 2023)
BiVO <sub>4</sub> /g-C <sub>3</sub> N <sub>4</sub> /NiFe <sub>2</sub> O <sub>4</sub>	OFX: 10 mg/L, 100 mL, pH = 9	300 W Xe lamp (λ > 420 nm)	100 mg	20 min, 93.8%	h <sup>+</sup> , •O <sub>2</sub> <sup>-</sup> , •OH, e <sup>-</sup> , <sup>1</sup> O <sub>2</sub>	(Zhao et al. 2021b)
LaFeO <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub> /BiFeO <sub>3</sub>	CIP: 10 mg/L, 50 mL, pH = 6.3	300 W Xe lamp	20 mg	60 min, 98.6%	•O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , •OH	(Saravanakumar and Park 2021)

<sup>a</sup> The main active species in each reference are shown boldly in the table

are discharged into aquatic and terrestrial ecosystems, which has become a serious problem. Table 3 lists the recent articles on g-C<sub>3</sub>N<sub>4</sub>-based materials in antibiotic degradation. As an economic and effective method to deal with antibiotic pollution, photocatalysis technology has attracted extensive attention.

The photocatalytic degradation efficiency of a single photocatalyst can be improved by changing the crystallinity, lattice vacancy, and other methods. For example, three g-C<sub>3</sub>N<sub>4</sub> with different crystallinity were prepared by thermal polymerization using urea, thiourea, and melamine as precursors (Phoon et al. 2022). The g-C<sub>3</sub>N<sub>4</sub> prepared from melamine, thiourea, and urea precursor were labeled MGCN, TGCN, and UGCN, respectively. According to the XRD analysis, the photocatalysts prepared with three precursors were successfully prepared. Compared with the other two materials, MGCN had higher crystallinity than the other two materials, because melamine formed an ordered melem intermediate, which was conducive to charge carrier migration. All of them were mesoporous materials with paralleled plate narrow pore structures. Due to abundant ammonia released by urea during thermal polymerization, UGCN had a larger surface area and a larger pore size than the other two samples. It can be seen from XPS that the C-S-C peak appeared in the C 1 s of the TGCN sample due to the presence of S in thiourea, suggesting that S was combined with the GCN structure. The authors also pointed out that the carbon/nitrogen ratio of MGCN was 0.697, while that of TGCN and UGCN were 0.650 and 0.668, respectively, which confirms the existence of N defects in MGCN. TGCN exhibited the strongest optical absorption capacity in the UV range, and MGCN showed better optical absorption than UGCN in 400–600 nm. The smallest band gap of TGCN was 2.61 eV, the MGCN was 2.63 eV, and the UGCN was 2.91 eV, indicating that the introduction of S atoms enhanced its light absorption ability. The CB of MGCN, TGCN, and UGCN was −0.33, −0.91, and −0.66 V (vs. NHE), the CB potential of MGCN was more negative than (O<sub>2</sub>/•O<sub>2</sub><sup>−</sup>), and the VB potential of MGCN was more positive than (H<sub>2</sub>O/•OH), indicating that during the photodegradation for MGCN, O<sub>2</sub> reduction and water oxidation can occur at the same time. However, the VB of TGCN and UGCN was more negative than the water oxidation potential, which meant that water oxidation would not happen. MGCN showed the highest photoactivity in the photocatalytic degradation of TC, because the dense structure promoted charge transfer and reduced the band gap. In addition, the presence of nitrogen vacancies prevented rapid charge recombination and promoted more charge migration on the surface of the MGCN. Therefore, it

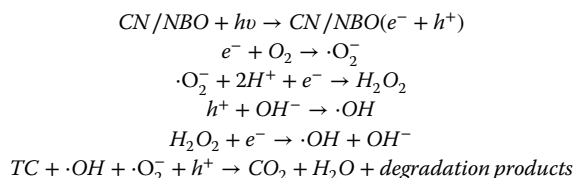
removed 99.5% of TC in 240 min, while the TC removal rates of TGCN and UGCN were only 85.1% and 94.3%. The photodegradation mechanism for MGCN was explored. In the presence of benzoquinone (BQ), ethylenediaminetetraacetic acid disodium (EDTA), and isopropyl alcohol (Jaleh et al. 2021), the degradation inhibition rates of TC were 84.8%, 74.8%, and 60.5%, respectively. Hence, the superoxide group was the most effective species to degrade TC. The degradation process was:



MGCN exhibited the strongest photocatalytic activity. This is because of its relatively small band gap and high crystallinity promoted visible light absorption and rapid migration of carriers. At the same time, the defects generated by nitrogen vacancies can effectively slow down charge recombination.

The built-in electric field generated by the formation of Z-type heterojunction in composite materials accelerates charge separation and improves the photocatalytic performance of photocatalysts. For example, nano flowers like NaBiO<sub>3</sub> (NBO) supported by 2D g-C<sub>3</sub>N<sub>4</sub> with Z-type heterojunction were prepared through a simple hydrothermal method (Wu et al. 2021). NaBiO<sub>3</sub>·2H<sub>2</sub>O and a certain amount of CN were added into the NaOH solution, and hydrothermally treated at 120 °C for 12 h. XRD results showed that the peak of NBO retained in the g-C<sub>3</sub>N<sub>4</sub>/NaBiO<sub>3</sub> (CN/NBO) photocatalyst, which indicated that g-C<sub>3</sub>N<sub>4</sub> did not affect the crystal structure of NBO. The morphology of composites studied by SEM showed that g-C<sub>3</sub>N<sub>4</sub> was a 2D nanosheet, while NBO was a 3D nanoflower. The smaller NBO was tightly combined on the surface of g-C<sub>3</sub>N<sub>4</sub> in the composite, which further confirmed the successful construction of CN/NBO heterostructure. Photocatalytic experiments were carried out on the prepared materials with tetracycline as the target pollutant. With the addition of the percentage of g-C<sub>3</sub>N<sub>4</sub> in the composite, the degradation efficiency of the composite first increased and then decreased. The degradation efficiency of the composite with 5% of CN was the highest. In addition, the photocatalyst still degraded 80% of TC in the eighth degradation. The photocatalyst used for 8 times was tested by XRD. The obvious characteristic peaks of CN and NBO could still be found in the sample after eight runs, which proved

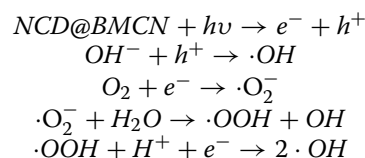
the stability of the composite material. To clarify the mechanism of photocatalytic degradation, IPA, p-benzoquinone (PBQ), and ethylenediaminetetraacetic acid disodium (EDTA-2Na) were used to test the active species in the reaction process. The degree of inhibition on tetracycline degradation was IPA > PBQ > EDTA-2Na, which showed that  $\cdot\text{OH}$  played a key role in the degradation process, followed by  $\cdot\text{O}_2^-$  and  $h^+$ . The corresponding reactions were as follows:



The built-in electric field of the Z-type heterojunction photocatalyst prepared by the hydrothermal method was conducive to accelerating charge separation and providing a higher redox potential for the carrier to form free radicals. Therefore, more active groups were formed on CN/NBO to degrade tetracycline, which provided an effective method for antibiotic degradation.

A similar method can also be used to prepare a ternary photocatalyst. For example, An indirect Z-type nitrogen-doped carbon dots (NCD) modified  $\text{Bi}_2\text{MoO}_6/\text{g-C}_3\text{N}_4$  ternary photocatalyst (NCD@BMCN) was prepared by hydrothermal method (Dang et al. 2021). NCD was obtained by hydrothermal treatment of the mixture of citric acid and Ethylenediamine solution at 160 °C for 4 h. Then,  $\text{g-C}_3\text{N}_4$  nanosheets (CN) were obtained by calcining urea at 500 °C for 2 h.  $\text{Bi}_2\text{MoO}_6/\text{g-C}_3\text{N}_4$  (BMCN) was synthesized by hydrothermal method.  $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$  and  $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$  were dissolved in diethylene glycol solution, then CN dispersed in ethanol was added. The mixture was hydrothermally reacted at 160 °C for 4 h to obtain a yellow BMCN precipitate. Finally, the prepared NCD solution was added to the BMCN suspension and stirred for 24 h. The prepared product was washed and vacuum dried to obtain NCD@BMCN. The 10–20 nm NCDs were uniformly deposited on the surface of BMCN in NCD@BMCN, which increased specific surface area, photocurrent density, and charge transfer efficiency. The existence of NCD on the hexagonal diffraction plane of the internal region of  $\text{sp}^2$  graphite carbon was confirmed from its fast Fourier transform (FFT) image, indicating that NCD particles,  $\text{Bi}_2\text{MoO}_6$  rods, and CN nanosheets were well combined to form heterostructures. The UV–Vis spectra showed that the absorption edge of BMCN had a red shift relative to pure  $\text{g-C}_3\text{N}_4$  due to the narrower band gap of  $\text{Bi}_2\text{MoO}_6$  compared with that of  $\text{g-C}_3\text{N}_4$ . The addition of NCD further expanded its visible light absorption range and significantly improved

the light absorption capacity. NCD@BMCN removed 99% of CIP under visible light in 30 min compared with BMCN (65%) and  $\text{g-C}_3\text{N}_4$  (20%). The photodegradation efficiency of CIP by different photocatalysts was simulated by pseudo-first-order equation. The pseudo-first-order rate constant of NCD@BMCN was 3.4 times and 15 times higher than that of BMCN and CN, respectively, which proved that NCD played a key role in improving photocatalytic efficiency. To reveal its photocatalytic degradation mechanism, tert-butanol (t-BuOH) and 1,4 benzoquinone (1,4-BQ) were added to the solution. The removal efficiency of CPX was significantly reduced, indicating that  $\cdot\text{OH}$  and  $\cdot\text{O}_2^-$  were the main active species. In addition, after the addition of ammonium oxalate monohydrate (AO), the photocatalytic efficiency did not decrease significantly, indicating that the  $h^+$  contributed little to the photocatalytic degradation. The degradation mechanism was as follows:



Under the visible light irradiation condition, the ternary photocatalyst showed good degradation ability for CPX, owing to the uniform distribution of BMO nanorods on the CN nanosheets and the increased specific surface area of the composite material. The NCD@BMCN showed a Z-scheme mechanism with photoinduced  $e^-$  moving from the CB of  $\text{Bi}_2\text{MoO}_6$  to the VB of CN, thereby generating more photogenerated carriers and enhancing the photocatalytic performance of CIP degradation.

Generally,  $\text{g-C}_3\text{N}_4$ -based photocatalysts have been widely used in antibiotic degradation. However, the degradation effect of a single  $\text{g-C}_3\text{N}_4$  photocatalyst on antibiotics is limited. By changing the crystallinity and introducing lattice vacancies of  $\text{g-C}_3\text{N}_4$ , its photocatalytic ability to degrade antibiotics can be improved by increasing carriers' mobility and slowing charges' recombination. Moreover, the internal electric field in  $\text{g-C}_3\text{N}_4$  heterostructure photocatalysts accelerates charge separation, thereby promoting antibiotic degradation. These methods effectively enhance the photocatalytic degradation abilities of  $\text{g-C}_3\text{N}_4$  for antibiotics and provide ideas for controlling antibiotic pollution.

### 3.3 Degradation of phenols

The applications of  $\text{g-C}_3\text{N}_4$ -based photocatalysts have also been proposed for phenolic degradation. Table 4 lists the recent applications of  $\text{g-C}_3\text{N}_4$ -based photocatalysts



**Table 4** Information on the degradation of phenols by g-C<sub>3</sub>N<sub>4</sub>-based photocatalyst<sup>a</sup>

Catalyst	Process conditions	Light source	Catalyst dosage	Performance	Active species	Ref
Fe <sub>2</sub> O <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub> @N-TiO <sub>2</sub>	BPA: 4.5 mg/L, 200 mL pH = 3	249 W Xe lamp	40 mm × 25 mm	40 min, 100%	<b>•OH</b> , •O <sub>2</sub> <sup>-</sup>	(Kong et al. 2020)
g-C <sub>3</sub> N <sub>4</sub> -ThA <sub>x</sub>	BPA: 10 mg/L	300 W Xe lamp (λ ≥ 420 nm)		100 min, 98.8%	<b>•O<sub>2</sub><sup>-</sup>, h<sup>+</sup></b> , •OH	(Ge et al. 2021)
In <sub>2</sub> O <sub>3</sub> /O doped g-C <sub>3</sub> N <sub>4</sub>	BPA: 50 mg/L, 50 mL pH = 5	Xe lamp (λ > 420 nm)	50 mg	180 min, 91%	<b>•O<sub>2</sub><sup>-</sup>, h<sup>+</sup></b> , •OH	(Uddin et al. 2021)
CeO <sub>2</sub> /g-C <sub>3</sub> N <sub>4</sub>	BPA: 10 mg/L, 100 mL	500 W xenon lamp	100 mg	150 min, 94.1%	<b>•OH</b> , •O <sub>2</sub> <sup>-</sup> , h <sup>+</sup>	(Zhao et al. 2021d)
S, K-doped g-C <sub>3</sub> N <sub>4</sub>	BPA: 10 mg/L, 50 mL	300 W lamp (λ > 420 nm)	15 mg	100 min, 64%	<b>•OH, h<sup>+</sup>, •O<sub>2</sub><sup>-</sup>, <sup>1</sup>O<sub>2</sub>, e<sup>-</sup></b>	(Xu et al. 2022)
CdSe-quantum dots coupled P doped-g-C <sub>3</sub> N <sub>4</sub>	2,4-DCP: 10 mg/L, 80 mL	300 W Xe lamp (λ > 420 nm)	50 mg	60 min, 78%	<b>•O<sub>2</sub><sup>-</sup>, h<sup>+</sup></b> , •OH	(Raziq et al. 2020)
BiFeO <sub>3</sub> -g-C <sub>3</sub> N <sub>4</sub> -WO <sub>3</sub>	2,4-DCP: 20 mg/L, 40 mL	300 W Xe lamp	100 mg	60 min, 63%	<b>•OH</b> , h <sup>+</sup> , e <sup>-</sup>	(Ali et al. 2020)
g-C <sub>3</sub> N <sub>4</sub> /8-quinolinolato iron(III) (Q <sub>3</sub> Fe)/H <sub>2</sub> O <sub>2</sub>	2,4-DCP: 30 mg/L	300 W lamp (λ > 400 nm)	20 mg	80 min, 90%	<b>•OH</b> , •O <sub>2</sub> <sup>-</sup>	(Feng et al. 2017b)
g-C <sub>3</sub> N <sub>4</sub> -CdS/Bi <sub>4</sub> O <sub>5</sub> I <sub>2</sub>	ACE: 3 mg/L, 50 mL	300 W Xe lamp	25 mg	25 min, 80%	<b>•O<sub>2</sub><sup>-</sup>, <sup>1</sup>O<sub>2</sub></b> , •OH	(Li et al. 2021a)
CeO <sub>2</sub> /I-K doped g-C <sub>3</sub> N <sub>4</sub>	ACE: 10 mg/L, 20 mL pH = 11	Eight 8 W visible light lamps (λ = 465 ± 40 nm)	40 mg	120 min, 98%	<b>•O<sub>2</sub><sup>-</sup>, h<sup>+</sup></b> , •OH	(Paragas et al. 2021)
g-C <sub>3</sub> N <sub>4</sub> /WO <sub>3</sub>	PNP: 10 mg/L, 50 mL	300 W Xe lamp	50 mg	90 min, 100%	<b>•O<sub>2</sub><sup>-</sup>, h<sup>+</sup></b> , •OH	(Zhang et al. 2021e)
α-Fe <sub>2</sub> O <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>	PNP: 389.5 mg/L, 100 mL pH = 3 H <sub>2</sub> O <sub>2</sub>	160 W incandescent lamp (400 < λ < 780 nm)	200 mg	120 min	<b>•OH</b> , •O <sub>2</sub> <sup>-</sup> , h <sup>+</sup>	(Sun et al. 2021a, b)
Zn <sub>1-1.5x</sub> Fe <sub>x</sub> S/g-C <sub>3</sub> N <sub>4</sub>	PNP: 10 mg/L, 250 mL	500 W lamp	200 mg	120 min, 96%	<b>•OH</b> , •O <sub>2</sub> <sup>-</sup> , h <sup>+</sup> , e <sup>-</sup>	(Wang et al. 2020)
g-C <sub>3</sub> N <sub>4</sub> /ZnO	4-CP: 9.4 mg/L, 100 mL	300 W lamp (λ > 420 nm)	100 mg	60 min, 95%	<b>•OH</b> , •O <sub>2</sub> <sup>-</sup> , h <sup>+</sup>	(Wang et al. 2017)
g-C <sub>3</sub> N <sub>4</sub> /perylene tetracarboxylic diimide -Br	4-CP: 5 mg/L	λ > 254 nm	50 mg	60 min, 100%	<b>•OH</b> , •O <sub>2</sub> <sup>-</sup> , h <sup>+</sup>	(Liu et al. 2021a)
[Mo <sub>7</sub> O <sub>24</sub> ] <sup>6-</sup> /g-C <sub>3</sub> N <sub>4</sub>	4-CP; BPA	Xe lamp (λ > 420 nm)		80 min, 100%; 30 min, 100%	<b>•O<sub>2</sub><sup>-</sup>, h<sup>+</sup></b> , •OH	(Zhang et al. 2022a)
g-C <sub>3</sub> N <sub>4</sub> -OH/Ov-TiO <sub>2</sub>	Phenol: 20 mg/L, 50 mL	300 W Xe lamp (λ ≥ 420 nm)	50 mg	220 min, 100%	<b>•OH</b> , •O <sub>2</sub> <sup>-</sup> , h <sup>+</sup>	(Ma et al. 2021a)
TiO <sub>2</sub> /B-doped g-C <sub>3</sub> N <sub>4</sub>	Phenol: 20 mg/L, 20 mL	250 W visible light	20 mg	120 min, 87%	<b>h<sup>+</sup>, •OH</b> , •O <sub>2</sub> <sup>-</sup> , e <sup>-</sup>	(Behera et al. 2021)
N doped g-C <sub>3</sub> N <sub>4</sub>	Phenol: 10 mg/L	300 W lamp (λ > 420 nm)	1 g/L	180 min, 70.1%	<b>•O<sub>2</sub><sup>-</sup>, h<sup>+</sup></b> , •OH	(Zhu and Zhou 2021)
Bi <sub>2</sub> Se <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>	Phenol: 100 mg/L, 15 mL	60 W LED lamp (λ > 400 nm)	50 mg	180 min	<b>•O<sub>2</sub><sup>-</sup>, •OH</b> , h <sup>+</sup>	(He et al. 2022a)

<sup>a</sup> The main active species in each reference are shown boldly in the table

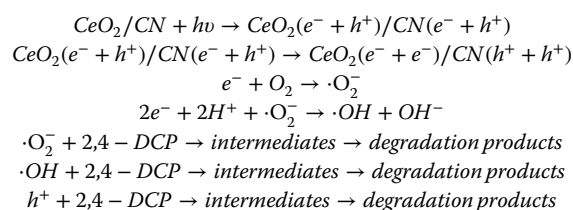
for phenolic removal. Modification of g-C<sub>3</sub>N<sub>4</sub> by doping and introducing defects are effective ways to improve the photocatalytic effect of g-C<sub>3</sub>N<sub>4</sub>. Modified g-C<sub>3</sub>N<sub>4</sub> was synthesized by thermal polymerization using urea as raw material and EDTA-2Na as a modifier to degrade BPA (He et al. 2022b). In XRD patterns, the (002) peaks of g-C<sub>3</sub>N<sub>4</sub> were shifted to lower angles and widened with

the increasing of EDTA-2Na addition, suggesting that the presence of EDTA-2Na slightly disturbed the in-plane structural packing of g-C<sub>3</sub>N<sub>4</sub>. Through FTIR spectra results, when the amount of EDTA-2Na reached a certain value, the signals of tri-s-triazine units and aromatic C-N heterocycles became small, confirming that the in-plane structure accumulation was disordered by EDTA-2Na.

The morphology of the materials was studied by SEM. The original g-C<sub>3</sub>N<sub>4</sub> showed a bulk and agglomerated structure, while modified g-C<sub>3</sub>N<sub>4</sub> showed a loose wrinkled structure, which might be due to the fact that the introduction of EDTA-2Na led to uneven distribution of electron density. In addition, the modified g-C<sub>3</sub>N<sub>4</sub> had a larger specific surface area, suggesting that it had more photocatalytic activity sites. On the other hand, XPS results showed that the presence of EDTA-2Na increased carbon content and might cause defects, suggesting that the presence of EDTA-2Na disturbed the planar structure of g-C<sub>3</sub>N<sub>4</sub>. Compared with g-C<sub>3</sub>N<sub>4</sub>, the edge of the absorption band of the modified g-C<sub>3</sub>N<sub>4</sub> sample showed an obvious red shift. In addition, with the increase of EDTA-2Na content, the doping of carbon in the g-C<sub>3</sub>N<sub>4</sub> framework led to the broadening of the absorption band, indicating that the absorption range of light expanded. At the same time, the band gap energy of g-C<sub>3</sub>N<sub>4</sub> was also reduced. In addition, modified g-C<sub>3</sub>N<sub>4</sub> impressively achieved 98.6% BPA photodegradation after 150 min when the original g-C<sub>3</sub>N<sub>4</sub> degraded only 39.6%. To clarify the strong photocatalytic ability of modified g-C<sub>3</sub>N<sub>4</sub> composites, the mechanism of charge transfer was presented. According to the characterization results, the combination of C doping and defects on g-C<sub>3</sub>N<sub>4</sub> can broaden the optical adsorption scope, improve the transfer of carriers, and reduce the recombination of e<sup>-</sup> and h<sup>+</sup>. Under optical irradiation, abundant photoinduced e<sup>-</sup> and h<sup>+</sup> were produced. The e<sup>-</sup> also moved to the surface and reacted with oxygen to form •O<sub>2</sub><sup>-</sup>. In addition, •O<sub>2</sub><sup>-</sup> and h<sup>+</sup> produced by modified g-C<sub>3</sub>N<sub>4</sub> played an important role in the photocatalytic degradation of BPA. C doping and defects reduced the band gap of g-C<sub>3</sub>N<sub>4</sub> and accelerated the migration of photogenerated carriers, which made the modified g-C<sub>3</sub>N<sub>4</sub> produce more excited electrons under visible light excitation and enhanced the separation ability of photogenerated electron holes, resulting in a significant enhancement of photocatalytic effect.

Combining g-C<sub>3</sub>N<sub>4</sub> with other materials is another effective method to improve the photocatalytic activity of g-C<sub>3</sub>N<sub>4</sub> in the degradation of phenols. For example, CeO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> composites were prepared using a wet chemical solution method (Humayun et al. 2019). g-C<sub>3</sub>N<sub>4</sub> was obtained by calcining dicyandiamide at 550 °C in the air for 2 h. The solution of cerium nitrate and ammonia was hydrothermally reacted at 160 °C for 12 h and further calcined at 500 °C for 2 h to obtain CeO<sub>2</sub>. Then, g-C<sub>3</sub>N<sub>4</sub> and a certain amount of CeO<sub>2</sub> were dispersed in a mixed solvent of ethanol and water, stirred and dried to remove the solvent. Finally, the dried product was annealed in air at 500 °C for 2 h to obtain CeO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> with different amounts of CeO<sub>2</sub>. XRD results showed that the diffraction peaks of CeO<sub>2</sub> appeared in the composites. With the

increase of CeO<sub>2</sub> content in composites, the peak intensity gradually increased and shifted to a larger direction of 2θ, indicating that g-C<sub>3</sub>N<sub>4</sub> and CeO<sub>2</sub> were successfully coupled. The SEM images showed that the small size CeO<sub>2</sub> nanoparticles were wrapped by layered g-C<sub>3</sub>N<sub>4</sub> irregular particles, and the particles became clearer as the amount of CeO<sub>2</sub> increased. The TEM results showed that CeO<sub>2</sub> particles were well dispersed on the surface of g-C<sub>3</sub>N<sub>4</sub>. In addition, the selective area electron diffraction images confirmed that CeO<sub>2</sub> existed in the form of nanocrystals. Therefore, a heterojunction was formed between CeO<sub>2</sub> and g-C<sub>3</sub>N<sub>4</sub>, which facilitated the migration of electrons between the two particles. The photocatalytic test of the composite material showed that g-C<sub>3</sub>N<sub>4</sub> degraded 29% and CeO<sub>2</sub> degraded 34% of 2,4-DCP after irradiation for 2 h, which was attributed to the small particle size with more active sites and higher specific surface area. After the coupling of the two, the photocatalytic effect was significantly improved. The composite material with 15% CeO<sub>2</sub> content had the best activity, which degraded 57% of 2,4-DCP in 2 h, while the photocatalytic efficiency of composite material with 20% CeO<sub>2</sub> content was slightly reduced. This is because the excessive CeO<sub>2</sub> acted as the recombination center of the charge carrier, resulting in a decrease in the photocatalytic effect. In addition, the cycle test showed that the photocatalytic activity of the composite did not change after 4 cycles, indicating that the photocatalyst had good stability. The CeO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> scavenger trapping test found that •OH was the main active material and the effects of •O<sub>2</sub><sup>-</sup> and h<sup>+</sup> were relatively small, and the degradation steps were as follows:



Compared with bare g-C<sub>3</sub>N<sub>4</sub>, the photocatalytic activity of the composite was significantly enhanced because the two components had a suitable energy platform that promoted the separation and migration of charge carriers.

The formation of double Z-type heterojunctions by preparing ternary compounds to promote the photocatalytic activity of photocatalysts has also attracted wide attention. A novel ternary g-C<sub>3</sub>N<sub>4</sub>/Bi<sub>2</sub>MoO<sub>6</sub>/CeO<sub>2</sub> (CBC) nanocomposite was prepared by solid-phase pyrolysis-assisted ultrasonic dispersion method for the degradation of 4-CP (Gao et al. 2022). The microstructure of the synthesized samples was observed by SEM and TEM. The original CeO<sub>2</sub> was spherical, and

$\text{Bi}_2\text{MoO}_6$  nanosheets were homogeneously wrapped on the surface of  $\text{CeO}_2$  microspheres. The  $\text{g-C}_3\text{N}_4$  had an ultra-thin layered structure after the two-step polymerization of melamine. Atomic force microscopy (AFM) analysis showed that the average thickness of the  $\text{g-C}_3\text{N}_4$  nanosheets was less than 5 nm. This structure can not only increase the surface active sites, but also expand the light response range and facilitate the transport of photogenerated carriers. The TEM images of CBC composites showed that the  $\text{Bi}_2\text{MoO}_6/\text{CeO}_2$  was successfully anchored on the  $\text{g-C}_3\text{N}_4$  and formed a close interfacial contact, which facilitated the effective transfer of photogenerated charge between different nanostructures. In addition, CBC exhibited a larger light response range. The CBC showed the highest degradation efficiency of 99.1% for 4-CP. A reaction mechanism was proposed for the enhanced photocatalytic performance. The  $e^-$  and  $h^+$  of  $\text{g-C}_3\text{N}_4$ ,  $\text{Bi}_2\text{MoO}_6$ , and  $\text{CeO}_2$  were generated on CB and VB, respectively. Combined with XPS analysis, the photoexcited  $e^-$  in the CB of  $\text{Bi}_2\text{MoO}_6$  can be transferred to the VB of  $\text{g-C}_3\text{N}_4$  and  $\text{CeO}_2$ , respectively, consuming the above  $h^+$  to achieve photogenerated carrier separation. The  $h^+$  on  $\text{Bi}_2\text{MoO}_6$  VB can directly degrade 4-CP, while the  $e^-$  left in CB of  $\text{g-C}_3\text{N}_4$  and  $\text{CeO}_2$  can quickly react with  $\text{O}_2$  to form  $\cdot\text{O}_2^-$ . In addition, some  $\cdot\text{O}_2^-$  was converted to  $\cdot\text{OH}$ . Finally, 4-CP was synergistically degraded by reactive radicals such as  $\cdot\text{O}_2^-$ ,  $h^+$ , and  $\cdot\text{OH}$ . Therefore, the ternary heterojunction and double Z-type charge transport mechanism were formed in the CBC nanocomposites. Through simple solid-state thermal decomposition assisted ultrasonic dispersion, the spherical  $\text{Bi}_2\text{MoO}_6/\text{CeO}_2$  was uniformly dispersed on the  $\text{g-C}_3\text{N}_4$  nanosheets, and a double Z-type heterojunction was formed by close interface contact, which realized the effective separation of electron-hole pairs and promoted the activity of the photocatalyst.

Thus,  $\text{g-C}_3\text{N}_4$ -based photocatalyst has been widely used in the degradation of phenolic pollutants. Under the light irradiation, the electrons on the VB migrate to the CB, and photogenerated electrons and holes will further react to generate active free radicals for phenolic pollutants degradation. In conclusion, introducing defects and preparation of Z-type heterojunction material are effectively strategies for improving the photodegradation ability of  $\text{g-C}_3\text{N}_4$  for phenolic pollutants.

#### 4 Conclusions and perspectives

In the present review, we highlight the applications of  $\text{g-C}_3\text{N}_4$  and its derivatives in the photodegradation of organic pollutants in water. The  $\text{g-C}_3\text{N}_4$ -based photocatalysts have the advantages of high efficiency, saving

energy, and reusability, making them promising photocatalysts for environmental applications, especially with respect to organic pollutant photodegradation. Further investigations thus should be conducted to promote research in this field and fill following important knowledge gaps. 1) Previous studies have mainly concentrated on improving the photodegradation activity of  $\text{g-C}_3\text{N}_4$ -based materials under laboratory experimental conditions. The degradation activity and mechanism for organic pollutants in the actual industrial wastewater are still unclear. 2) The applicability of  $\text{g-C}_3\text{N}_4$  photodegradation technique under natural conditions needs to be further evaluated. 3) Obstacles have yet to be addressed for the upscale from laboratory to commercially available photocatalytic wastewater treatment technology. 4) Most of the relevant laboratory studies have only focused on the removal of one pollutant at a time. However, it rarely happens in real wastewater, which usually contains various pollutants including both easily degradable and recalcitrant ones.

#### Abbreviations

4-CP	4-Chlorophenol
4-NP	4-Nitrophenol
2,4-DCP	2,4-Dichlorophenol
ACE	Acetaminophen
AO7	Acid Orange 7
BPA	Bisphenol A
CPX	Ciprofloxacin
CQDs	Carbon quantum dots
CR	Congo red
MB	Methylene blue
MG	Malachite green
MO	Methyl orange
NOR	Norfloxacin
OFX	Ofloxacin
OTC	Oxytetracycline
PNP	P-nitrophenol
RhB	Rhodamine B
SMX	Sulfamethoxazole
TC	Tetracycline
ThA	3-Thiophenemalonic acid

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The data are available from the corresponding author on reasonable request.

## Declarations

### Competing interests

The authors declare no competing interests.

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