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Recent advances on g-C₃N₄-based Z-scheme photocatalysts for organic pollutant removal

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Graphitic carbon nitride (g-C₃N₄)-based Z-scheme photocatalysts have attracted growing interest in organic pollution removal because of their strong light absorption capacity, abundant active sites, and outstanding photocatalytic performance. This review provides a survey of the latest advances of q-C₃N₄based Z-scheme photocatalysts for organic pollution removal, beginning with the fundamentals of g- $C_{\overline{z}}N_4$ -based Z-scheme photocatalysts. Subsequently, the characterization strategies and preparation approaches for g-C₃N₄-based Z-scheme photocatalysts are discussed. Various typical g-C₃N₄-based Z-scheme photocatalysts for organic pollutant degradation are described. Finally, this work provides some new insights into the major challenges, favorable circumstances, heuristic perspectives, and future development direction.

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1. Introduction

Water pollution has attracted growing interest because of the rapid industrial development and expanding population. 1-6 The exploration and development of advanced technologies,

 $adsorption, ^{7-10} \\$ electrocatalysis, 11,12 such photocatalysis 13-19 (especially visible-light-responsive photocatalysis), 20-24 are very promising for overcoming these issues. Among them, the most promising strategy is photocatalysis because of its unique advantage in simply using inexhaustible solar energy to organic pollution removal. 19,25-31 Graphitic carbon nitride (g-C₃N₄)-based materials, as one of the most up-and-coming photocatalysts, have gained dramatically growing interests in organic pollution removal owing to their easy functionalization, band electronic and structure, physicochemical stability and photocatalytic property.32-47 Although great progress has been made, the photocatalytic performance of single g-C₃N₄ is still far from practical



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production demand owing to the fast electron-hole (e⁻-h⁺) recombination and poor light absorption capacity. 48-51 Hence, it is crucial to explore new g-C₃N₄-based photocatalysts with strong light absorption capacity and high e⁻-h⁺ separation for organic pollutant removal.



Mini review

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approaches have been proposed, e.g., doping, 52,53 obtaining porous architectures, 54,55 achieving highly crystalline loading, 57,58 structures,56 metal heterojunction construction, 59-61 exfoliation, 62,63 defect engineering, 64,65 and molecular and electronic structure engineering. 66-69 Among these approaches, the integration of two semiconductors to fabricate type-II heterojunctions is a promising method for enhancing the photocatalytic performance since the band alignment between the two semiconductors will provide high separation efficiency of the photogenerated carrier. 70,71 However, their photocatalytic activity is not significantly enhanced because of electrostatic repulsion.⁷² To overcome this disadvantage, an emerging g-C₃N₄-based Z-scheme system was proposed by the Yu group⁷³ that created a more negative conduction band (CB) and more positive valence band (VB), and further achieved stronger photocatalytic ability. 74,75 When compared to type-II heterojunctions, the charge migration of Z-scheme systems is more favorable. Owing to their strong light absorption capacity, abundant active sites, and outstanding photocatalytic performance, ⁷⁶ g-C₃N₄-based Z-scheme photocatalysts have achieved increasing attention for organic pollutant removal.

To enhance the photocatalytic property of g-C₃N₄, various



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Although there have been many inspired reviews on gphotocatalysts C₃N₄-based for various applications, 32,33,71,77-88 a comprehensive review about g-C₃N₄-based Z-scheme photocatalysts for organic pollutant removal is nonexistent despite the great interest in this research topic. In this review, the fundamentals of g-C₃N₄based Z-scheme photocatalysts are firstly discussed. Subsequently, the characterization strategies and preparation methods for g-C₃N₄-based Z-scheme photocatalysts are illustrated. Next, some typical g-C3N4-based Z-scheme photocatalysts for organic pollutant removal are described. Finally, this work provides some advanced insights into the major challenges, favorable circumstances, heuristic perspectives, and future development direction.

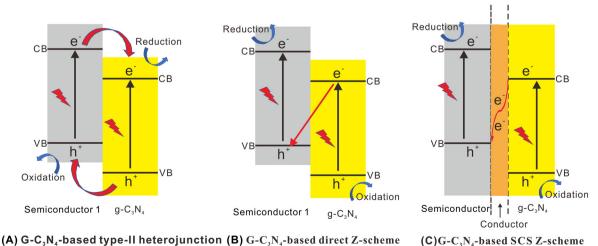
2. What are $g-C_3N_4$ -based Z-scheme photocatalysts?

g-C₃N₄-based Before introducing the photocatalysts, it is crucial to fully comprehend g-C₃N₄-based type-II heterojunctions, e.g., CeO₂/g-C₃N₄,⁸⁹ Co₃O₄/g-C₃N₄,⁹⁰ Cu₂O/g-C₃N₄, ⁹¹ Fe₂O₃/g-C₃N₄, ⁹² g-C₃N₄/g-C₃N₄, ⁹³ h-BN/flowerring g-C₃N₄,⁹⁴ which display outstanding photocatalytic property owing to their high separation efficiency of photogenerated charges through the band alignment.74,95 In these type-II heterojunctions, both CB and VB potentials of semiconductor 1 are lower than those of g-C₃N₄ (Fig. 1A). The e in the CB of semiconductor 1 have a tendency to migrate to the CB of g-C₃N₄ owing to the band alignment. Similarly, the h⁺ in the VB of g-C₃N₄ easily migrate to the VB of semiconductor 1.96,97 Owing to the e and h being concentrated on g-C₃N₄ and semiconductor 1, respectively, high separation efficiency of the photogenerated e⁻ and h⁺ can be achieved through type-II heterojunctions. 32,72,98,99 However, there are still some potential problems that need to be solved. The photocatalytic activity of the charges are not

significantly enhanced. 100,101 It is also very difficult for the ein semiconductor 1 and h⁺ in g-C₃N₄ to migrate to the CB of g-C₃N₄ because of strong electrostatic repulsions, such as e⁻e or h+h interactions, respectively. Thus, it is critical to fabricate a new type of photocatalytic system to relieve this disadvantage.

Inspired by bionic photosynthesis, there is great potential in the development of the Z-scheme photocatalysts for enhancing photocatalytic performance. 71,102 In 2013, the Yu group reported on the first use of a direct Z-scheme mechanism to explain the outstanding photocatalytic formaldehyde (HCHO) oxidation performance over TiO₂/g-C₃N₄ hybrids.⁷³ Especially, the direct Z-scheme systems exhibit a structure similar to that of type-II heterojunctions (Fig. 1A), but they possess an evidently dissimilar charge migration mechanism. 103,104 The charge migration path of the direct Z-scheme photocatalysts resembles the letter "Z" (Fig. 1B), in which the e in g-C₃N₄ with stronger oxidation ability will integrate with the h+ in semiconductor 1 with strong reduction ability. 105 Therefore, the e in semiconductor 1 with stronger reduction ability and h⁺ in g-C₃N₄ with stronger oxidation ability may be retained. As a result, the photocatalytic property of the direct Z-scheme systems can be evidently enhanced. Moreover, the charge migration path of the direct Z-scheme systems is more favorable than that of type-II heterojunctions. This is because the migration of the photogenerated e from the CB of g-C₃N₄ to the VB of semiconductor 1 with adequate photogenerated h⁺ is very favorable owing to the effective electrostatic attraction.72 It is believed that g-C3N4-based Z-scheme systems exhibit faster charge transfer¹⁰¹ when compared with g-C₃N₄-based type-II heterojunctions.

Until now, there have been two main types of g-C₃N₄based Z-scheme systems, including g-C₃N₄-based direct Z-scheme (Fig. 1B) without an electron mediator and g-C₃N₄-based semiconductor-conductor-semiconductor (SCS)



(C)G-C₃N₄-based SCS Z-scheme

Fig. 1 Comparative analysis of the charge transfer mechanism on g-C₃N₄-based type-II heterojunctions (A), g-C₃N₄-based Z-scheme (B) and (C) g-C₃N₄-based SCS Z-scheme systems.

Z-scheme system (Fig. 1C) with an electron mediator (conductor), e.g., conductive polymer, graphene, noble metal. The g-C₃N₄-based direct Z-scheme systems (e.g., TiO₂/g-C₃N₄,⁷³ ZnO/g-C₃N₄,¹⁰⁸ WO₃/g-C₃N₄,¹⁰⁹ Ag₃PO₄/ g-C₃N₄,¹¹⁰ BiVO₄/g-C₃N₄,¹¹¹ Bi₂WO₆/g-C₃N₄, ¹¹² BiOCl/g-C₃N₄, ¹¹³ AgBr/g-C₃N₄ (ref. 114)) display outstanding photocatalytic performance by keeping a very negative CB and a very positive VB. 115 As for g-C₃N₄-based SCS Z-scheme systems (e.g., WO₃/Cu/gC₃N₄, ¹¹⁶ Ag₃PO₄/Ag/g-C₃N₄, ¹¹⁷ CdS/Au/g-C₃N₄ (ref. 106)), a conductor acting as a mediator is utilized to link two components for more carriers. 118 migration of photogenerated favorable Moreover, the p-n Z-scheme systems (e.g., C₃N₄-Pdg-C₃N₄/CaFe₂O₄, 121 Cu₂O, 119 $LaFeO_3/g-C_3N_4$, ¹²⁰ phosphorus nanosheets/FeSe₂/g-C₃N₄ (ref. 122)) favorably formed by combining p-type and n-type materials have received increasing attention. In the comprehensive consideration of the photocatalytic activity and fabrication cost, we must compare the advantages and disadvantages of the two g-C₃N₄-based Z-scheme systems, and develop a more appropriate photocatalytic system to achieve optimal photocatalytic property. The balance of charge utilization is also vital. 123 The consumption of e- and h+ for redox reactions should be unified. Otherwise, the accumulation of charges may lead to the self-oxidation/reduction of the photocatalysts or the mediators. 124

3. Characterization strategies for g-C₃N₄-based Z-scheme photocatalysts

As shown above, the Z-scheme systems exhibit similar structures with type-II heterojunctions. Therefore, it is important to prove that the charge migration mechanism occurs via multifarious characterization strategies, e.g., selfconfirmation via photocatalytic reactions, products, and radical species, selective photodeposition of noble metal, X-ray photoelectron spectroscopy (XPS), surface photovoltage (SPV) techniques, density function theory (DFT) calculation, and others.

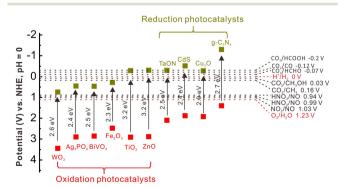


Fig. 2 Standard redox potentials of some commonly semiconductors. Reproduced with permission from ref. 35.

3.1. Self-confirmation *via* photocatalytic reactions, products, and radical species

It is generally known that not all photogenerated charges centered on the photocatalysts' surface are utilized for the photocatalytic reaction. Typically, there are only some photogenerated e in a semiconductor with enough redox potential that can be utilized for photocatalytic reactions. 76,125,126 Some commonly used standard redox potentials are shown in Fig. 2. Z-scheme systems can be confirmed via standard redox potentials and investigating the photocatalytic reactions, products, and radical species.

Considering the g-C₃N₄/WO₃ photocatalysts as an example, a tight contact between g-C₃N₄ and WO₃ can be found (Fig. 3A), and the individual g-C₃N₄ and WO₃ components exhibit low photocatalytic performance (Fig. 3B and C). Interestingly, the g-C₃N₄/WO₃ composites enhanced photocatalytic performance. phenomenon is mainly attributed to the fabrication of a direct Z-scheme system (Fig. 3D), which accelerated the separation of photogenerated e⁻-h⁺. The g-C₃N₄/WO₃ composites also exhibited good stability (Fig. 3E).127 If it follows type II mechanism, the photogenerated e will transfer from the CB of g-C₃N₄ to the CB of WO₃. In this way, the g-C₃N₄/WO₃ composites would display reduced redox capability, which conflicts with the experimental results. Therefore, it should obey the Z-scheme direct mechanism.

We can also prove the Z-scheme mechanism through analyzing the photocatalytic products. Tao and co-workers 128 proved the successful fabrication of the g-C₃N₄/CoTiO₃ direct heterojunctions through analyzing photocatalytic products. The single CoTiO₃ displayed very low photocatalytic activity due to its low reduction ability, but the single g-C₃N₄ exhibited moderate photocatalytic performance owing to its adequate reduction ability. After the successful construction of a direct Z-scheme photocatalyst, the photogenerated carrier separation efficiency and the redox ability of g-C₃N₄/CoTiO₃ were evidently improved. If the migration route of the photogenerated carriers follows the type-II mechanism, the photogenerated e would be focused on CoTiO3 without adequate reduction potential, which would result in very low photocatalytic activity. Nevertheless, the experimental results unveiled that g-C₃N₄/CoTiO₃ exhibited enhanced photocatalytic property, suggesting that the g-C₃N₄/CoTiO₃ composites should follow the Z-scheme direct mechanism. This work demonstrated that the photocatalytic tests feasibly study the charge migration mechanism.

3.2. Selective photodeposition of noble metals

As for Z-scheme systems, the photogenerated charges can be efficiently separated. Photodeposition of several noble metals, e.g., Au, Ag, Cu, on electron-rich areas is a promising strategy to unlock the photocatalytic reaction

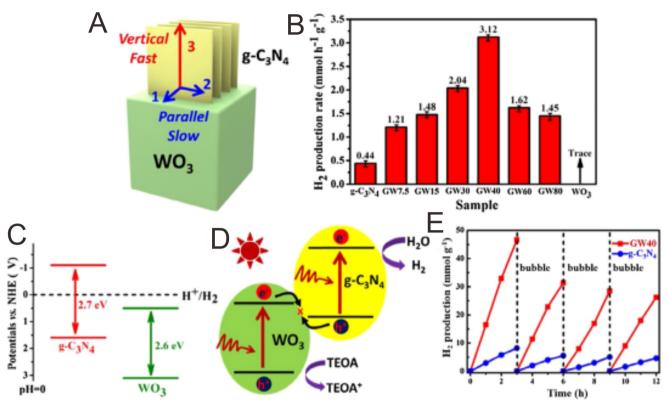


Fig. 3 (A) The growth and extension of $g-C_3N_4$ parallel with and perpendicular to the flat surface of the WO₃ nanocuboids. (B) Comparison of the photocatalytic H_2 production properties. (C) Band structure of $g-C_3N_4$ and WO_3 . (D) The direct Z-scheme charge migration path in the $g-C_3N_4$ WO₃ composites. (E) The photocatalytic stability tests of the g-C₃N₄ and g-C₃N₄/WO₃ composites. Reproduced with permission from ref. 127.

sites. 129 On the basis of the photodeposition sites of noble metals, the charge migration route and Z-scheme mechanism can be further proved. For example, Yu and co-workers¹³⁰ proposed Fe₂O₃/g-C₃N₄ photocatalysts, which showed improved photocatalytic performance when compared with the pure Fe₂O₃ and g-C₃N₄ components. The TEM image (Fig. 4) showed that Pt nanoparticles were deposited on g-C₃N₄, but not on Fe₂O₃, suggesting that the photogenerated e mainly focused on g-C₃N₄. If there were no close contacts between g-C₃N₄ and Fe₂O₃, Pt nanoparticles would randomly photodeposit on g-C₃N₄ and

Fe₂O₃. If it followed the type II mechanism, the Pt nanoparticles would mainly concentrate on Fe₂O₃. Instead, the Pt nanoparticles mainly concentrated on g-C₃N₄. Therefore, the Fe₂O₃/g-C₃N₄ systems should follow the Z-scheme mechanism.

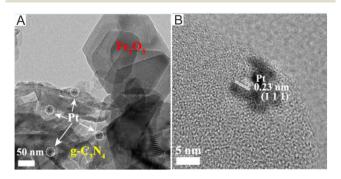


Fig. 4 (A) TEM image and (B) HRTEM image of the Fe₂O₃/q-C₃N₄ composites after photodeposition of Pt nanoparticles. Reproduced with permission from ref. 130.

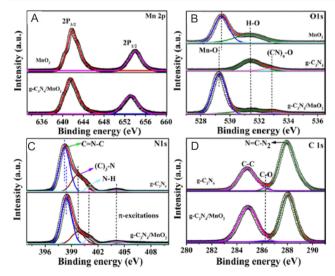


Fig. 5 XPS spectra of (A) Mn 2p, (B) O 1s, (C) N 1s, and (D) C 1s for g-C₃N₄, MnO₂, and g-C₃N₄/MnO₂. Reproduced with permission from ref.

3.3. XPS characterization

XPS is a promising technique for analyzing the photocatalysts' surface chemistry, 9,131-135 and it is also used for investigating the transformations in the electronic density on the photocatalysts' surface via probing the shift in the binding energies. 98,136 A positive shift indicates a decrease of the electron density. Therefore, a shift in the binding energy can be used to study the migration path of photogenerated e. For g-C₃N₄/MnO₂, 137 the Mn 2p spectrum of the MnO₂ nanosheets (Fig. 5A) displayed two main peaks located at 641.9 and 654.0 eV, corresponding to Mn $2P_{3/2}$ and Mn $2P_{1/2}$, respectively, which shifted to 641.6 and 653.7 eV. However, the pure g-C₃N₄ sample only exhibited the adsorbed oxygen species and other oxygen impurities (Fig. 5B). In view of g-C₃N₄/MnO₂, a combined O 1s spectrum was obtained (Fig. 5B) with the binding energy of the lattice oxygen shifted to 529.3 eV. The N 1s spectrum of g-C₃N₄ (Fig. 5C) can be divided into three main peaks at 398.2, 399.7, and 401.2 eV, which can be assigned to the sp²-hybridized nitrogen (C=N-C), bridging nitrogen ((C)₃-N), and N-H bonding in the g-C₃N₄ framework, respectively. When compared with the MnO₂ nanosheets, the N 1s binding energy of the C=N-C bond displayed a minor shift to higher value (Fig. 5D). These results suggested the formation of a Z-scheme system through the coating of MnO2 onto the g-C3N4 nanosheets with electrons flowing from g-C₃N₄ to MnO₂ and achieving thermodynamic equilibrium. The XPS valence band spectra

are also used for calculating band edge potentials, which can further explain the Z-scheme mechanism. 138

3.4. SPV techniques

Recently, the SPV strategy has provided some new opportunities to study the charge transfer path of Z-scheme systems. 139 Typically, Kelvin probe force microscopy (KPFM) can easily obtain the photocatalysts' surface potential distribution with high spatial resolution and potential sensitivity. 140-143 Therefore, using KPFM-based techniques to study the carrier transfer path of Z-scheme systems is an efficient technology. Guan and co-workers¹⁴⁴ reported a g-C₃N₄-based Z-scheme system through encapsulation of g-C₃N₄ into hollow In₂O₃ tubules. To gain a deeper understanding of the charge migration route, KPFM was proposed (Fig. 6A-D). Under dark conditions, the difference in potential between the sample surface and substrate surface was 51.87 mV. Conversely, under visible light illumination, the surface potential of the whole area is greatly reduced, suggesting the concentration photogenerated e and h. Meanwhile, the difference in potential between the sample surface and substrate surface were decreased to 50.57 mV. This change confirmed that the photogenerated e migrated from the surface of g-C₃N₄ to the interior of In₂O₃, further illustrating the Z-scheme mechanism. Therefore, the SPV strategy is also very promising for the confirmation of the Z-scheme mechanism.

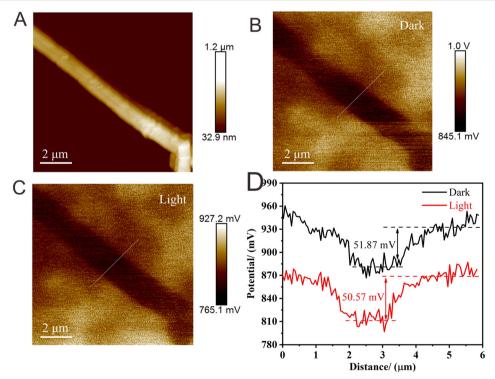


Fig. 6 (A) Atomic force microscopy image of the samples. (B) KPFM potential image of the samples under dark conditions. (C) KPFM potential image of the samples under light illumination. (D) The corresponding surface curves correspond to the white light. Reproduced with permission from ref. 144.

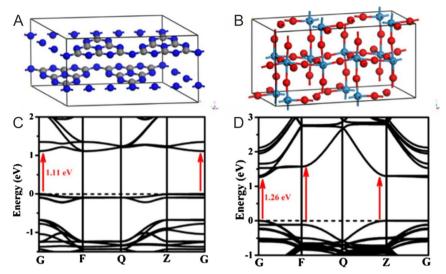


Fig. 7 Crystal structures of (A) g-C₃N₄ and (B) monoclinic WO₃. The calculated electronic band structures of (C) g-C₃N₄ and (D) WO₃. Reproduced with permission from ref. 127.

3.5. DFT calculation

It is well known that DFT calculation is a promising technology to explore the charge migration mechanism. 145,146 In detail, the first-principles simulation with a DFT calculation is commonly utilized for studying the charge transfer through analyzing the effective mass of its charge to estimate the recombination efficiency of the photogenerated carrier. 108,127,147 The DFT calculation is conducted based on the crystal and electronic band structures (Fig. 7).127 The effective masses of the charges of WO3 and g-C3N4 can be calculated through the parabolic fitting of their CB minimum and VB maximum based on eqn (1) and (2), respectively, and the separation efficiency of the photogenerated h⁺ to the e⁻ based on eqn (3):

$$m^* = \hbar^2 (d^2 E/dk^2)^{-1} \tag{1}$$

$$v = \hbar k/m^* \tag{2}$$

$$D = m_h^*/m_e^* \tag{3}$$

where \hbar , d^2E/dk^2 , k, m^* , ν , D, m_h^* , and m_e^* respectively signify the reduced Planck constant, the coefficient of the secondorder term in a quadratic fit of E(k) curves for the band edge,

Table 1 Calculated effective masses of the charges of g-C₃N₄ and WO₃

Species	Effective mass	<i>G–F</i> direction	<i>G–Z</i> direction	F–Q direction	Z–Q direction
g-C ₃ N ₄	$m_{ m h}^*$	0.72	71	_	
	m_{e}^{*}	0.53	1.8		
	D	1.36	39.44	_	_
WO_3	$m_{ m h}^*$	0.53	_	0.49	0.51
	$m_{ m e}^*$	0.16	_	0.17	0.14
	D	3.31	_	2.88	3.64

the wave vector, the effective mass of a charge, the transfer efficiency of a charge, separation efficiency of the photogenerated h⁺ to the e⁻, and the effective mass of photogenerated h⁺ and e⁻.

The calculated effective masses of photogenerated e and h⁺ and the relevant separation efficiency in G-Z and G-F directions are summarized in Table 1. The smaller D value for $g-C_3N_4$ (D = 1.36) indicates its faster recombination efficiency. In contrast to g-C₃N₄, the excited electrons of WO₃ exhibit a higher transfer efficiency from bulk to interface, and then transfer into g-C₃N₄. In this way, the interfacial electron transfer path in g-C₃N₄/WO₃ occurs from WO₃ to g-C₃N₄, which follows the direct Z-scheme mechanism. 127

4. Preparation methods for $q-C_3N_4$ based Z-scheme photocatalysts

Until now, there have been a lot of approaches utilized for the preparation of g-C₃N₄-based Z-scheme photocatalysts, and selecting a suitable method is crucial. In the following sections, different preparation methods for g-C₃N₄-based Z-scheme photocatalysts are described. Their advantages and disadvantages are also discussed in Table 2.

4.1. Deposition-precipitation strategy

Deposition-precipitation strategy is regarded as an effective method fabricating g-C₃N₄-based Z-scheme photocatalysts. 148 Narayanan and co-workers reported on a AgCl@g-C3N4 direct Z-scheme photocatalyst through a modified deposition-precipitation method. 149 AgNO3 was firstly introduced into the g-C₃N₄ suspension, and then vigorously stirred to ensure full adsorption of Ag+ on the g-C₃N₄ surface. Subsequently, the Cl⁻ ions and Ag⁺ ions were fully coated on the g-C₃N₄ surface, and then reacted to form

Table 2 Comparison between different methods for the preparation of Z-scheme photocatalysts

Synthesis strategy	Merits	Demerits
Deposition–precipitation strategy	High-efficiency, simple, and low-cost	Uncontrollable morphology and easy aggregation
Hydrothermal and solvothermal strategy	(i) Facile, eco-friendly, and highly efficient (ii) Produced photocatalysts with a relatively high crystallinity, narrow size distribution, and controllable morphology	Complicated preparation process
Solid-state synthesis	High-efficiency, simple, and low-cost	Uncontrollable morphology and easy aggregation
Ion-exchange strategy	The formation of closely contact interface, and the initial morphology can be well preserved	Time- and cost-consuming and complicated preparation process
Electrospinning strategy	Very suitable for the preparation of 1D materials	Time- and cost-consuming, complicated preparation process, and low efficiency
Self-assembly strategy	Narrow size distribution and uniform morphology	Time- and cost-consuming, and complicated preparation process
Photo-deposition strategy	Facile, eco-friendly, and highly efficient	Easy aggregation
Mechanical agitation strategy	Facile, eco-friendly, and highly efficient, and reduced aggregation	Relatively poor interface contacts between the two semiconductors

the AgCl@g-C₃N₄ direct Z-scheme photocatalysts. The coating of AgCl on g-C₃N₄ was proved via XRD and FTIR spectra, which also confirmed the close interface between AgCl and g-C₃N₄ for greatly enhancing the photocatalytic performance.

4.2. Hydrothermal and solvothermal strategy

Hydrothermal and solvothermal approaches are usually conducted in a sealed autoclave under high temperature and pressure conditions, endowing the obtain semiconductors with high crystallinity, regulatable morphology, homogeneous size distribution. 13,150-155 Therefore, this strategy is commonly utilized for fabricating g-C₃N₄-based Z-scheme photocatalysts without further post-treatment. Liu and co-workers¹⁵⁶ reported on Bi₂WO₆ quantum dots/g-C₃N₄ binary Z-scheme systems via a simple one-pot hydrothermal strategy. In this work, porous g-C₃N₄ ultrathin nanosheets provided a high specific surface area for decorating Bi₂WO₆ quantum dots, leading to a strong interaction between the Bi₂WO₆ quantum dots and g-C₃N₄ nanosheets.

4.3. Solid-state strategy

The solid-state strategy is commonly defined as the calcination of one precursor or multiple precursors in a gas atmosphere at an elevated temperature. 157,158 homogeneous dispersion of the precursors plays an important role in affecting the quality and uniformity of the prepared materials. Several methods, such as intensive ball milling¹⁵⁹ and dissolving precursors in a solvent followed by drying,160 can evidently stop the agglomeration of the precursors and maintain uniformity. The calcination parameters, such as heating rate and calcination temperature and time, can regulate the crystallinity, microstructure morphology, surface performance, and phase structure, which further affect the photocatalytic performance. Wang and co-workers¹⁶¹ explored the effect of calcination temperature on the preparation of hierarchical TiO₂/g-C₃N₄ Z-scheme photocatalysts. The insertion

nanoparticles on the porous TiO2 framework was confirmed via SEM and XRD (Fig. 8A-G). The BET results indicated the effect of the calcination temperature on the porous structure (Fig. 8H and I). The specific surface area and pore volume gradually increased with increasing temperature to 550 °C, but a further increase in the temperature would lead to a reduction.

4.4. Ion exchange strategy

The ion exchange method mainly involves exchanging ions in an ionic crystal, but the framework still keeps close intact with each other. 162-164 Additionally, the ion exchange can be quickly accomplished when the temperature is raised. Wang and co-workers¹⁶⁵ prepared Cd_xZn_{1-x}S/Au/g-C₃N₄ Z-scheme photocatalysts through the ion exchange approach, which displayed improved photocatalytic H2 production activity. Furthermore, when the ion exchange strategy is used, it is much easier to control the final product via adjusting the reaction conditions. Additionally, the Janus-like structures ensured them with improved visible light absorption and enlarged absorption range. Thus, the ion exchange method is an efficient approach for post-modification of Z-scheme systems.

4.5. Electrospinning strategy

It is well known that the electrospinning method is a versatile strategy to fabricate 1D materials, e.g., nanofibers, nanotubes, nanobelts. 166,167 It can also be utilized for the preparation of 1D Z-scheme photocatalysts. A 1D nanostructure can boost the mobility of charge and enhance the absorption of visible light, and thus decrease the recombination of charge and enhance the photocatalytic performance. 168,169 Li and co-workers 170 prepared a TiO2/g-C₃N₄/RGO ternary Z-scheme heterojunction via an electrospinning coupled approach. The crystal structure, morphology, composition, specific surface



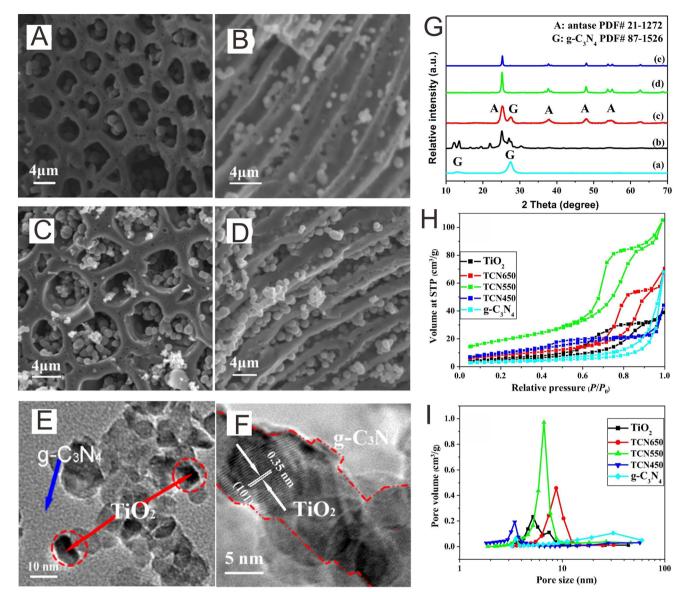


Fig. 8 SEM images at different perspective views of (A and B) the pure hierarchical TiO₂ framework and (C and D) the TiO₂/g-C₃N₄ Z-scheme photocatalyst calcination at 550 °C (denoted as TCN550). TEM (E) and HRTEM (F) images of the TCN550. XRD (G), N2 adsorption isotherms (H), and pore size distribution (I) for the different samples. Reproduced with permission from ref. 161.

photoelectrochemical properties, and photocatalytic performance can be adjusted *via* electrospinning condition.

4.6. Self-assembly strategy

The self-assembly approach involves the spontaneous organization of a disordered component into an ordered structure and the optimization of the free energy.8,171-174 Cheng and co-workers¹⁷⁵ proposed a simple electrostatic selfassembly approach for preparing g-C₃N₄/ZnO Z-scheme photocatalysts. In this system, the electrostatic force between g-C₃N and ZnO was very strong, which can concurrently enhance the charge separation and stability. When N-doped ZnO nanoplates were added to the exfoliated g-C₃N₄ suspension, the ZnO nanoplates' surface was spontaneously coated with g-C₃N₄ nanosheets to minimize the surface energy, resulting in the formation of a core-shell N-doped ZnO/g-C₃N₄ Z-scheme photocatalyst. Therefore, the selfassembly method endows the Z-scheme photocatalysts with a narrow size distribution, regulatable morphology, and strong interaction.

4.7. Photo-deposition strategy

The photo-deposition method is the phenomenon in which a semiconductor is decorated on the other's surface via light illumination. 177-179 In this method, the CB potential of the semiconductor must be much lower than the reduction potential, and the VB potential must be higher than the oxidation potential. 129 The photo-deposition method is very

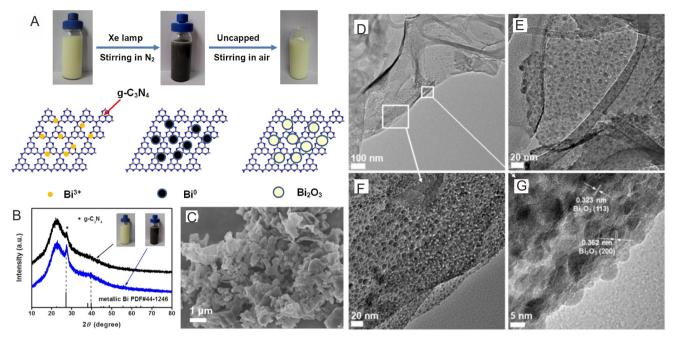


Fig. 9 (A) Diagrammatic sketch of the preparation of Bi₂O₃/g-C₃N₄. (B) XRD patterns of the intermediate products. (C) FESEM image of Bi₂O₃/g-C₃N₄. C₃N₄. TEM (D-F) and HRTEM (G) images of Bi₂O₃/g-C₃N₄. Reproduced with permission from ref. 180.

promising for preparing Z-scheme photocatalysts. A Bi₂O₃/g-C₃N₄ direct Z-scheme photocatalyst was fabricated through the simple photoreductive deposition of Bi3+ on the g-C3N4 surface under light illumination, followed by air oxidation of Bi³⁺ to Bi₂O₃ (Fig. 9A). The uniform dispersion of quantum-sized Bi₂O₃ on the g-C₃N₄ nanosheet surface could be observed from the XRD pattern, FESEM, TEM and HRTEM images (Fig. 9B-G). Wu and co-workers 181 reported a novel BiVO₄(040)-Ag@CdS Z-scheme photocatalyst in which Ag was selectively photo-deposited on the (040) facets of BiVO₄, and the CdS nanorods were decorated on BiVO₄(040)-Ag. Thus, the photo-deposition strategy is also a promising technology decorating one semiconductor with the other semiconductor, which further improves the charge separation.

4.8. Mechanical agitation approach

The conventional mechanical agitation strategy involves the attachment of two pre-prepared semiconductors via physical methods, such as grinding, 182 impregnation, 183 planetary ball-milling,184 and reflux.185 However, the semiconductors assembled through these methods have no close interface, and they are easily separated from each other, resulting in a low charge transfer efficiency and moderate photocatalytic performance.¹⁸⁶ Ohno and coworkers¹⁸⁷ fabricated three kinds of g-C₃N₄/WO₃ Z-scheme photocatalysts via grinding, impregnation assisted with sonication, and planetary ballmilling methods. Different from the agglomerated WO₃ decorated on the g-C₃N₄ nanoplate prepared through the first two methods, WO₃ prepared through planetary ball-milling was uniformly dispersed on the $g-C_3N_4$

nanoparticles, which was beneficial for improving the specific surface area and photocatalytic performance.

Among the various fabrication strategies, depositionprecipitation, hydrothermal and solvothermal, solid-state synthesis, ion-exchange, electrospinning, self-assembly, and photo-deposition strategies are beneficial for achieving a strong interaction and intimate interface between the two semiconductors, which will lead to improved charge migration efficiency. 76,188 The strong interaction (e.g., covalent bond, ionic bond, and coordination bond) and wear interaction (hydrogen bond and van der Waals force) can be formed between the two semiconductors, serving as charge transfer channels and enhancing the interface contact. Especially, the ion-exchange strategy is very promising for forming a close contact interface, and the initial morphology can be retained. 189,190 The self-assembly method can ensure the two semiconductors with a narrow size distribution and uniform morphology. 191 In contrast, the mechanical agitation method exhibits relatively poor interface contact between the two semiconductors, resulting in low photocatalytic performance. 192 Nevertheless, this strategy is simple and cost effective, and shows great potential in practical production (Table 2).

5. Photocatalytic organic pollutant degradation

Many organic pollutants, such as methylene blue (MB), methyl orange (MO), 4-nitrophenol (4-NP), rhodamine B (RhB), basic fuchsin (BF), bisphenol A (BPA), brilliant blue 2,4-dichlorophenol (2,4-DCP), tetracycline (TC), sulfamethoxazole (SMX), indomethacin (IDM), and others

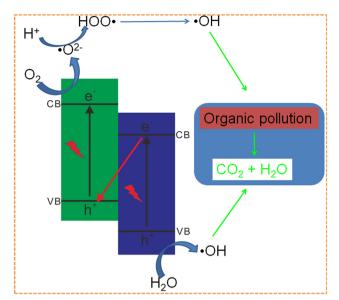


Fig. 10 Diagrammatic explanation of organic pollutant degradation over q-C₃N₄-based Z-scheme photocatalysts.

have recently been excessively discharged to soil and water, which have negative influences on the environment and human health. 193-201 g-C₃N₄-based Z-scheme photocatalysts have gained growing attention for organic pollutant removal because of their enhanced photocatalytic efficiency and stability (Fig. 10).77,101,202 Some important examples for photocatalytic organic pollutant degradation are shown below, and their property comparison is shown in Table 3.

5.1. g-C₃N₄/TiO₂ Z-scheme photocatalysts

TiO2 is valuable in the photocatalytic fields because of its unique advantages, e.g., excellent controllability, large surface area, amazing stability, and ease of recycling. 203,204 Some investigations have focused on combining TiO2 with g-C3N4 to develop Z-scheme photocatalysts for achieving better photocatalytic property. The first example was reported by Yu and co-workers,73 who achieved markedly enhanced by $g-C_3N_4/TiO_2$ photocatalytic properties Z-scheme photocatalysts. Dong and co-workers²⁰⁵ fabricated an allsolid-state (ASS) Z-scheme photocatalyst through decorating g-C₃N₄ nanosheets on TiO₂ nanotube arrays (TNTAs). The asprepared photocatalyst displayed improved photocatalytic property for RhB degradation, which was about 4 times greater than that exhibited by pure TiO2 TNTAs. Ho and coworkers²⁰⁶ prepared the g-C₃N₄ quantum dots (QDs)-modified rutile TiO₂ (rTiO₂) (g-C₃N₄ QDs-rTiO₂) hybrid via calcination of the mixture of P25 TiO2 and melamine under 500 °C for 4 h, which displayed excellent photocatalytic properties for the RhB degradation. Facet engineering is crucial for improving the photocatalytic performance. The exposed {010} and {110} crystal facets of BiVO4 have been demonstrated to be the active reduction and oxidation sites, respectively.207 Raj and co-workers²⁰⁸ fabricated a g-C₃N₄/TiO₂/CuO Z-scheme heterostructure via a simple hydrothermal strategy, which displayed improved photocatalytic performance of (90.3%) RhB dye degradation within 120 min. In addition, almost no loss in photocatalytic activity after 5 cycles was found. The Lv group²⁰⁹ studied the influence of the exposed {001} and {101} facets of TiO2 on the photocatalytic activity. They first fabricated TiO2 nanosheets (NSs) and TiO2 nanoboxes (NBs), and then combined them with g-C₃N₄ to prepare TiO₂ NS/g-C₃N₄ and TiO₂ NBs/g-C₃N₄, respectively (Fig. 11A and B). The {001} facets of TiO2 NSs came into contact with g-C3N4 (Fig. 11A and C), and the {101} facets of TiO₂ NBs came into contact with g-C₃N₄ (Fig. 11B and D). Under visible light illumination, the photogenerated carriers on TiO2 were separately accumulated on the {101} and {001} facets. 210 The photogenerated electrons were subsequently centered on the {101} facets of the TiO2 NBs, which could transfer to g-C3N4 for photocatalytic reactions, which is in line with the direct Z-scheme mechanism (Fig. 11F). Thus, the e-h⁺ separation efficiency of TiO2 NBs/g-C3N4 is much higher than that of TiO2 NSs/g-C3N4, and the TiO2 NBs/g-C3N4 displayed the highest photocatalytic efficiency (Fig. 11E). This work offers us a new design concept, wherein the suitable interface engineering of the exposed facets of a pure semiconductor in a direct Z-scheme photocatalyst is able to effectively enhance the e-h+ separation, which can further improve the photocatalytic property.

5.2. g-C₃N₄/WO₃ Z-scheme photocatalysts

WO3 is gradually becoming recognized as one of the most promising photocatalysts, owing to its outstanding photocatalytic performance, chemical inertness, and optical stability.211,212 In comparison with TiO2, WO3 possesses a smaller band gap (2.7 eV) and more positive CB level. Thus, it is also favorable for combining with g-C₃N₄ to construct Z-scheme photocatalysts. The Chen group²¹³ developed a WO₃/g-C₃N₄ Z-scheme photocatalyst. The reaction rate constants for degrading MB and BF over WO₃(5.0 wt%)/g-C₃N₄ were found to be 4.77 and 3.84 times greater than that of the bare g-C₃N₄, respectively. The Jia group²¹⁴ developed a new bifunctional WO₃/g-C₃N₄ Z-scheme photocatalyst by in situ liquid phase strategy. The photocatalytic degradation efficiency of the 30 wt%WO₃/g-C₃N₄ was found to be 2.5 and 2.7 times higher than the individual g-C₃N₄ and WO₃, respectively. Kang and co-workers²¹⁵ fabricated WO₃/g-C₃N₄ Z-scheme photocatalysts through a simple one-step simultaneous heating approach utilizing urea as the starting material, which exhibited markedly improved photocatalytic property for MB degradation. Chai and co-workers²¹⁶ constructed a WO₃/g-C₃N₄ Z-scheme photocatalyst through in situ acidic precipitation and subsequent calcination processes, which also showed improved photocatalytic property for RhB degradation. Zheng and co-workers²¹⁷ reported a C-doped g-C₃N₄/WO₃ Z-scheme photocatalyst through the hydrothermal strategy. Benefiting from high specific areas, C-substitution, and Z-scheme mechanism, the

 $\textbf{Table 3} \quad \text{Comparison of the photocatalytic organic pollution degradation performance over g-C_3N_4-based Z-scheme photocatalysts g_{3}-g_{4}$-g_{5}$$

Photocatalysts	Preparation strategy	Mass (mg)	Light source	Target pollutant/initial concentration	Degradation time (min) and rate (%)	$\frac{K_{\text{app}}^{a}}{\left[10^{-2} \text{ min}^{-1}\right]}$	Ref
g-C ₃ N ₄ /TiO ₂	Calcination route	300	15 W UV lamp, 365 nm	HCHO/170 ppm	60 (~94)	7.36	73
g-C ₃ N ₄ /TiO ₂	Electrochemical route	_	300 W xenon lamp, >420 nm	RhB/5 mg L ⁻¹	300 (~77)	4.48	205
g-C ₃ N ₄ /rTiO ₂	Calcination route	50	500 W xenon lamp, >420 nm	RhB/5 mg L ⁻¹	240 (~98)	11.5	206
g-C ₃ N ₄ /TiO ₂ /CuO	Hydrothermal strategy	50	500 W xenon lamp	$RhB/0.5 \text{ mg L}^{-1}$	120 (~97)	0.95	208
g-C ₃ N ₄ /TiO ₂ -HNB	Microwave-assisted	_	3 W LED lamp,	Brilliant red X-3B/1.0	45 (~99)	5.1	209
	strategy		365 nm	\times 10 ⁻⁴ mmol L ⁻¹			
WO_3/g - C_3N_4	Ball milling and heat	10	500 W xenon	MB and BF/0.9 × 10^{-5}	MB 120 (~99) BF	_	213
	treatment methods		lamp, >470 nm	and $1.0 \times 10^{-5} \text{ mol L}^{-1}$	120 (~97)		
WO_3/g - C_3N_4	In situ liquid phase process	50	300 W xenon lamp, >400 nm	$MB/50 \text{ mg L}^{-1}$	90 (~97)	1.90	214
WO_3/g - C_3N_4	One-step heating procedure	30	500 W xenon lamp, >420 nm	RhB/5 mg L^{-1}	120 (~90)	_	215
C-doped g-C ₃ N ₄ /WO ₃	Cellulose-assisted	100	500 W xenon	TC/10 mg L ⁻¹	60 (~70)	_	217
C doped g C3114/1103	strategy	100	lamp, >420 nm	10/10 mg L	00 (70)		217
WO ₃ /g-C ₃ N ₄ /Bi ₂ O ₃	One step co-calcination	100	300 W xenon	$TC/10 \text{ mg L}^{-1}$	60 (~80)	2.37	159
00 0 1 2 0	way		lamp, >420 nm	5	,		
g-C ₃ N ₄ /MnO ₂	Wet-chemical method	50	300 W xenon lamp	$RhB/10 mg L^{-1}$	180 (~77)	3.3	137
Bi_2O_3/g - C_3N_4	Ball milling and heat	5	500 W xenon	MB and RhB/1.1 \times 10 ⁻⁵	MB 120 (~76)	2.53 and	223
	treatment methods		lamp, 470 nm	and $1.0 \times 10^{-5} \text{ mol L}^{-1}$	RhB 120 (~73)	1.01	
g-C ₃ N ₄ /Bi ₂ O ₃ /BiPO ₄	Calcination route	15	_	$MO/10 \text{ mg L}^{-1}$	160 (~88)	1.15	225
β -Bi ₂ O ₃ /g-C ₃ N ₄	Hydrothermal	40	350 W xenon	$RhB/1 \times 10^{-5} mol/L$	80 (~97)	4.22	226
	calcination approach		lamp, >420 nm				
g - C_3N_4/Bi_4O_7	Calcination method	80	500 W halogen lamp, 420–800 nm	MB and RhB/— and —	90 (~80)	1.73	227
g-C ₃ N ₄ /Bi ₂ WO ₆	Mixing and heating	50	300 W xenon	$^{2,4}\text{-DCP/20 mg L}^{-1}$	120 (>99)	1.13	228
8 -3 -4 2·····	method		lamp, 350-780 nm	,	(
g-C ₃ N ₄ /Bi ₂ WO ₆	One-pot hydrothermal strategy	50	Xenon lamp (XL-300, Yirda)	RhB/10 mg L^{-1}	30 (>99)	8.17	156
g-C ₃ N ₄ /RGO/Bi ₂ WO ₆	Hydrothermal method	250	500 W xenon	$TCP/20-100 \text{ mg L}^{-1}$	120 (>99)	3.15	229
g 03114/100/10121106	Tryarociterina incensa	200	lamp, >420 nm	101/20 100 Hig E	120 (> 33)	0.10	223
Ag-decorated	Polymerization method	40	300 W xenon	RhB/10 mg L^{-1}	90 (>99)	_	230
g-C ₃ N ₄ /Bi ₂ WO ₆	i olymendadon memod	10	lamp, 400 nm	ruib, ro mg b	30 (2 33)		
Bi_3O_4Cl/g - C_3N_4	Solid-phase calcination method	50	250 W xenon lamp, >420 nm	RhB and TC/10 and 10 ${\rm mg~L}^{-1}$	RhB 60 (>98) TC 60 (>76)	— and 2.05	231
BiOBr/RGO/pg-C ₃ N ₄	Solvothermal method	50	300 W xenon	RhB and TC/10 and 10	RhB 7.5 (>99) TC	— and —	232
blobl/RGO/pg-C ₃ N ₄	301votnermai metriou	30	lamp, 420 nm	$mg L^{-1}$	60 (>97)	— and —	232
$\mathrm{Bi}_{12}\mathrm{GeO}_{20}/\mathrm{g}\text{-}\mathrm{C}_3\mathrm{N}_4$	Sol-gel method	300	300 W Dy lamp,	RhB/5 mg L ⁻¹	50 (>99)	2.55	233
G 37 /GOP: 34 O	** 1 .1 .1.1	400	400 nm	0.37 1.1 1/45 7=1	4=0 (- 50)	0.53	
g-C ₃ N ₄ /C@Bi ₂ MoO ₆	Hydrothermal deposition method	100	500 W xenon lamp, >420 nm	β -Naphthol/15 mg L ⁻¹	150 (>62)	0.63	234
$Ag_3PO_4/g-C_3N_4$	In situ precipitation	5	300 W xenon	SMX/1 mg L^{-1}	10 (~98)	6.3	238
11631 04/6 03114	method	3	lamp, >400 nm	SWIN, I HIG E	10 (30)	0.3	250
Ag_3PO_4/g - C_3N_4	In situ precipitation method	30	300 W xenon lamp	$\mathrm{MO/10}~\mathrm{mg}~\mathrm{L}^{-1}$	5 (>99)	_	239
g-C ₃ N ₄ /Ag ₂ WO	In situ precipitation	30	300 W xenon lamp	$MO/10 \text{ mg L}^{-1}$	150 (~98)	_	240
	method						
$Ag_2CrO_4/g-C_3N_4$	Precipitation method	20	300 W xenon lamp, >400 nm	MO and RhB/10 and 10 mg L ⁻¹	MO 21 (~90) RhB 40 (~97)	9.64 and 9.21	241
Ag@AgBr/g-C ₃ N ₄	Deposition-precipitation	100		MO and MB/10 and 10	MO 10 (~97) RhB	— and —	117
Acr CrO /Acr/cr C N	method <i>In situ</i> growth and	F0	FOO III won on	mg L ⁻¹ 2,4-DCP/10 mg L ⁻¹	10 (~96)	1.50	0.42
$Ag_2CrO_4/Ag/g-C_3N_4$	photoreduction methods	50	500 W xenon	2,4-DCP/10 IIIg L	120 (~96)	1.52	243
g-C ₃ N ₄ /Ag/MoS ₂	In situ growth and	100	lamp, <420 nm 300 W xenon	RhB/20 mg L ⁻¹	60 (>99)		247
g-C ₃ N ₄ /Ag/N1O ₂	precipitation methods	100	lamp, >420 nm	KIIB/20 IIIg L	00 (>99)		24/
g-C ₃ N ₄ /ZnO	Ultrasonically exfoliated	100	300 W xenon	$4\text{-CP/}10^{-4} \; \text{mol L}^{-1}$	90 (>99)	5.09	248
G la/pag/ C 27	method	20	lamp, >420 nm	Aim to d	200 (02)	0.74	2.15
CdS/RGO/g-C ₃ N ₄	Hydrothermal method	20	350 W xenon lamp		300 (~92)	0.71	249
g-C ₃ N ₄ -RGO-TiO ₂	Liquid-precipitation	50	300 W xenon	RhB/30 mg L ⁻¹	180 (~92)	1.37	250
8 - 3 - 4 2	strategy		lamp, >420 nm				
$MoO_3/g-C_3N_4$	Calcination method	30	350 W xenon	$TC/20 \text{ mg L}^{-1}$	90 (~90)	2.23	251
		30	350 W xenon lamp, >420 nm	TC/20 mg L ⁻¹	90 (~90)	2.23	251

Table 3 (continued)

Photocatalysts	Preparation strategy	Mass (mg)	Light source	Target pollutant/initial concentration	Degradation time (min) and rate (%)	K_{app}^{a} [10 ⁻² min ⁻¹]	Ref.
NiTiO ₃ /g-C ₃ N ₄	Calcination method	40	30 W visible LED	RhB/10 mg L ⁻¹	_	3.09	253
LaFeO ₃ /g-C ₃ N ₄	Calcination method	30	300 W xenon lamp, >420 nm	RhB/25 mg L ⁻¹	60 (~98)	6.22	120
V_2O_5/g - C_3N_4	In situ growth strategy	50	250 W xenon lamp, 420 nm	RhB and TC/10 and 10 $\rm mg~L^{-1}$	120 (~77)	4.91 and —	107
$SnO_{2x}/g-C_3N_4$	Calcination method	100	350 W xenon lamp, >420 nm	$RhB/10 \text{ mg L}^{-1}$	40 (>99)	1	254
$InVO_4/Bi_2S_3/g\text{-}C_3N_4$	Wet-impregnation method	200	300 W xenon lamp, >420 nm	RhB19/15 mg L ⁻¹	100 (~96)	0.57	255
$CDs/g\text{-}C_3N_4/SnO_2$	Thermal polymerization	10	300 W xenon lamp, >420 nm	$IDM/10 \text{ mg L}^{-1}$	40 (>99)	2.33	256
$g\text{-}C_3N_4/ZnFe_2O_4/UiO\text{-}66$	Hydrothermal method	_	300 W xenon lamp, >420 nm	RhB/—	40 (>99)	4.89	256
$g\text{-}\mathrm{C}_3\mathrm{N}_4/\mathrm{BiVO}_4/\mathrm{CoFe}_2\mathrm{O}_4$	High-temperature solid-state route	50	300 W xenon lamp, >400 nm	RhB/—	25 (>99)	2.84	257
$g\text{-}C_3N_4/g\text{-}C_3N_4$	Bottom-up approach	10	50 W xenon lamp, >400 nm	RhB/10 mg L ⁻¹	240 (>99)	1.39	246

^a K_{app} represents the reaction rate constant.

prepared Z-scheme photocatalysts displayed a narrow bandgap, and improved visible-light absorption and separation efficiency of charge. All of the abovementioned improved photocatalytic properties of the WO₃/g-C₃N₄ composites were mainly because of the unusual merits of the Z-scheme photocatalytic systems.

5.3. g-C₃N₄/MnO₂ Z-scheme photocatalysts

MnO2 is gradually becoming recognized as an efficient photocatalyst owing to its excellent photocatalytic performance, suitable bandgap value, and optical stability.²¹⁸ Xu and co-workers¹³⁷ prepared g-C₃N₄/MnO₂ Z-scheme photocatalysts. There were many functional groups and exposed lone pair electrons from N atoms on the g-C₃N₄ surfaces, which provided abundant adsorption sites for Mn²⁺ ions (Fig. 12A). It should be noted that the thicknesses of the g-C₃N₄ nanosheets, MnO₂ nanosheets, and the g-C₃N₄/MnO₂ hybrid were approximately 3.4, 2.2, and 5.7 nm, respectively, which were roughly equal to the stacking distance of the g-C₃N₄ and MnO₂ nanosheets (Fig. 12B-D). The electrostatic potentials of MnO₂ (001) and g-C₃N₄ (001) were measured to be 6.8 and 4.5 eV, respectively, suggesting that the electrons on the g-C₃N₄ will spill onto MnO₂ via the interface (Fig. 12E-G). Thus, g-C₃N₄ was more positively charged. Photocatalytic experiments suggested that the prepared g-C₃N₄/MnO₂ exhibited better photocatalytic property when compared with the bare g-C₃N₄ and MnO₂ (Fig. 12H and I). Herein, electronic paramagnetic resonance (EPR) analysis can be used to survey the reactive species. The results indicated that the transfer of the photogenerated carrier in g-C₃N₄/ MnO₂ should obey the direct Z-scheme mechanism (Fig. 12J and K). Moreover, the positive slopes of the Mott-Schottky plots indicated that both g-C₃N₄ and MnO₂ are typical n-type semiconductors (Fig. 12J and K). With regard to n-type semiconductors, the flat-band potential is nearly that same as that for CB, 221,222 and the CB of g-C3N4 and MnO2 were measured to be -1.61 and 1.22 V, respectively (Fig. 12L and N). The VB positions were calculated by XPS spectroscopy (Fig. 12M), and the results showed that the VB of g-C₃N₄ and MnO₂ were about 1.81 and 3.26 V, respectively. Therefore, a Z-scheme transfer mechanism can be used to illustrate the enhanced photocatalytic property over the g-C₃N₄/MnO₂ hybrids. As for the individual g-C₃N₄ or MnO₂, the photogenerated h⁺ in g-C₃N₄ cannot oxidize OH⁻ to produce 'OH radicals and the photogenerated e in MnO2 cannot generate 'O2- radicals, respectively, because of the thermodynamic equilibrium limit. After the Z-scheme heterojunction was formed, the photogenerated e in the CB of MnO₂ will migrate to the VB of g-C₃N₄ and then combine with the h⁺. The Z-scheme configuration can explain the enhanced photocatalytic property well (Fig. 13O).

5.4. g-C₃N₄/bismuth oxide Z-scheme photocatalysts

Recently, multifarious bismuth-based semiconductors, e.g., O_4Cl , 231 BiOBr, 232 Bi $_{12}$ GeO $_2$, 233 Bi $_2$ MoO $_6$, 234 have been reported for combining with g-C₃N₄ to form Z-scheme photocatalysts because of their unique characteristics, such as outstanding electrical property, appropriate energy band positions, highly efficient photocatalytic performance. 235-237 For example, Chen and co-workers 223 prepared a Z-scheme Bi₂O₃/g-C₃N₄ photocatalyst. The reaction rate constants of Bi₂O₃ (1.0 wt%)/g-C₃N₄ for MB and RhB degradation were approximately 3.4 and 5 times greater that of the bare g-C₃N₄ and Bi₂O₃, respectively. The Z-scheme mechanism was confirmed through various technologies (e.g., ESR, PL-TA and PL, etc.), which resulted in enhanced photocatalytic property. Du and co-workers²²⁷ fabricated a

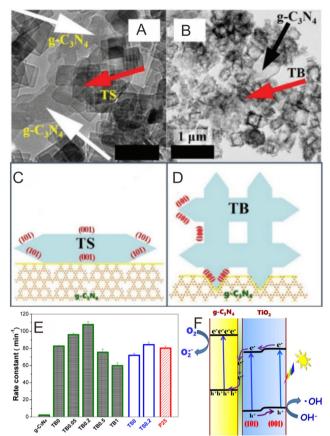


Fig. 11 TEM images of (A) TiO₂ NSs/g-C₃N₄ and (B) TiO₂ NBs/g-C₃N₄. Diagrammatic sketch of the contact interface between TiO2 and g- C_3N_4 on (C) TiO_2 $NSs/g-C_3N_4$ and (D) TiO_2 $NBs/g-C_3N_4$. (E) Comparison of the photocatalytic activity of the synthesized photocatalysts for the photocatalytic degradation of brilliant red X3B. (F) Diagrammatic sketch for the photoinduced charge carrier migration mechanism in the TiO₂/g-C₃N₄ hybrids. Reproduced with permission from ref. 209.

high-performance g-C₃N₄/Bi₄O₇ Z-scheme photocatalyst via a simple way. In contrast to the single photocatalysts, the asprepared g-C₃N₄/Bi₄O₇ photocatalyst displayed enhanced degradation efficiency toward MB, phenol, RhB, and BPA. The improved photocatalytic performance should be attributed to the improved light utilization and the highly efficient Z-scheme charge migration. Zhao and co-workers²²⁸ reported a simple mixing and heating strategy to fabricate the g-C₃N₄/Bi₂WO₆ Z-scheme photocatalyst. The optimal dechlorination rate constant of 2,4-DCP over g-C₃N₄/Bi₂WO₆ was 1.13 h⁻¹, exceeding those of the bare g-C₃N₄ and Bi2WO₆ by 7.6 and 8.9 times, respectively. Yin and co-workers²³³ prepared a new ASS $Bi_{12}GeO_{20}/g-C_3N_4$ Z-scheme photocatalyst. The obtained Bi₁₂GeO₂₀/g-C₃N₄ photocatalyst displayed improved photocatalytic property for degrading microcystin-LR and RhB in contrast to the individual Bi12-GeO₂₀ and g-C₃N₄. The enhanced photocatalytic property is because of the enhanced light utilization efficiency and effective spatial separation of the photo-generated carriers. In short, bismuth-based semiconductors can couple well with gC₃N₄ to fabricate Z-scheme photocatalysts for enhancing photocatalytic properties owing to their suitable CB and CB

5.5. g-C₃N₄/Ag-containing semiconductor Z-scheme photocatalysts

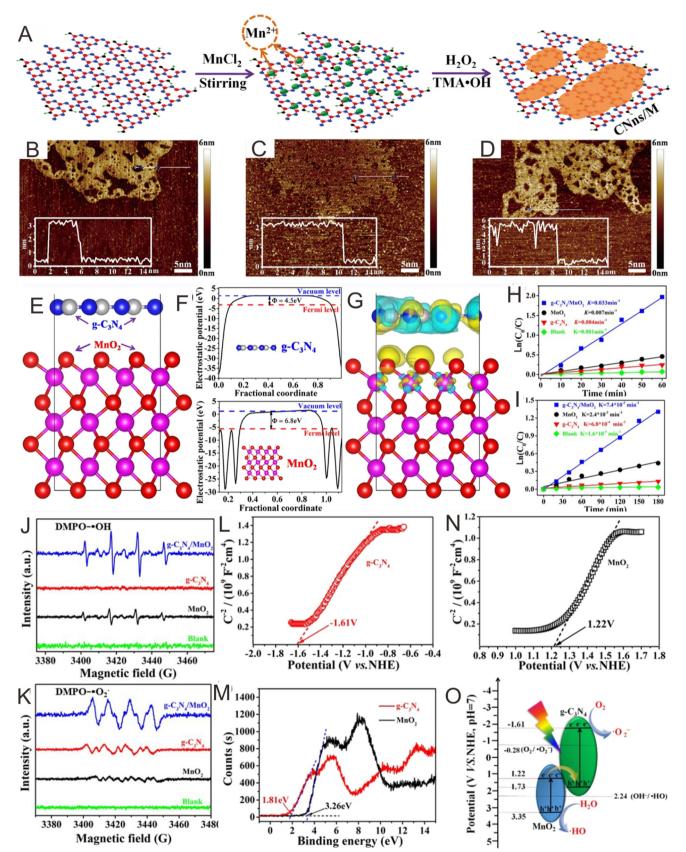
Ag-containing photocatalysts can greatly improve the photocatalytic property of g-C₃N₄ through the Z-scheme mechanism. Some examples of Ag-containing photocatalysts with improved properties include Ag₃PO₄/g-C₃N₄, ^{238,239} Ag₂-WO₄/g-C₃N₄,²⁴⁰ and Ag₂CrO₄/g-C₃N₄.²⁴¹ In particular, utilizing noble metal Ag as the electron mediator for fabrication of Agsemiconductors/g-C₃N₄ SCS photocatalysts can achieve more effective photocatalytic performance.²⁴² The Guo group¹¹⁷ fabricated various Ag@AgBr/g-C₃N₄ Z-scheme photocatalysts photoreduction of AgBr/g-C₃N₄ hybrids. A dramatically improved photocatalytic property of Ag@AgBr/g-C₃N₄ was achieved, and is mainly attributed to the high light utilization and Z-scheme mechanism. Quan and coworkers²⁴³ prepared Ag₂CrO₄/Ag/g-C₃N₄ Z-scheme photocatalysts via simple in situ growth and photoreduction strategies. Because of the strong light utilization, effective separation of the photogenerated e⁻ and h⁺, and the excellent redox ability, the obtained Ag₂CrO₄/Ag/g-C₃N₄ hybrids showed remarkably improved photocatalytic performance for 2,4-DCP degradation, exceeding that of the pure g-C₃N₄ by 5.2 times.

5.6. Metal-free g-C₃N₄-based Z-scheme photocatalysts

Fabricating metal-free g-C₃N₄-based Z-scheme photocatalysts is a very promising and environmentally friendly method for degrading organic pollutants.^{244,245} Huang and co-workers²⁴⁶ constructed a metal-free 2D g-C₃N₄/g-C₃N₄ Z-scheme photocatalyst via utilization of a bottom-up approach. Profiting from the 2D morphology with the large exposed active site and the Z-scheme mechanism with the efficient separation of the photoinduced carriers, the fabricated 2D g-Z-scheme photocatalysts C_3N_4/g - C_3N_4 showed photocatalytic performance when compared to the conventional g-C₃N₄/g-C₃N₄ homojunction and bulk g-C₃N₄. This work provides some new design concepts for Z-scheme systems with specific 2D morphology for enhanced organic pollutant removal.

5.7. g-C₃N₄-based SCS Z-scheme photocatalysts

As mentioned above, the g-C₃N₄-based SCS Z-scheme photocatalysts seem to be more promising for organic pollutant degradation. Among them, conductors acting as intermediates are utilized to link two semiconductors for the more favorable migration of the photogenerated carriers.33 A typical example was presented by the Fang group.²⁴⁷ They prepared a Z-scheme g-C₃N₄/Ag/MoS₂ ternary plasmonic Z-scheme photocatalyst with flowerlike nanostructures with a diameter of $\sim 0.4 - 0.6$ The prepared g-C₃N₄/Ag/MoS₂



 $\textbf{Fig. 12} \hspace{0.2cm} \textbf{(A)} \hspace{0.2cm} \textbf{Diagrammatic} \hspace{0.2cm} \textbf{sketch} \hspace{0.2cm} \textbf{of} \hspace{0.2cm} \textbf{g-C}_3\textbf{N}_4/\textbf{MnO}_2 \hspace{0.2cm} \textbf{prepared} \hspace{0.2cm} \textbf{through} \hspace{0.2cm} \textbf{\textit{in}} \hspace{0.2cm} \textbf{\textit{situ}} \hspace{0.2cm} \textbf{\textit{growth}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{MnO}}_2 \hspace{0.2cm} \textbf{\textit{on}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{g-C}}_3\textbf{N}_4 \hspace{0.2cm} \textbf{\textit{nanosheets.}} \hspace{0.2cm} \textbf{\textit{AFM}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{g-C}}_3\textbf{\textit{N}}_4 \hspace{0.2cm} \textbf{\textit{nanosheets.}} \hspace{0.2cm} \textbf{\textit{AFM}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{g-C}}_3\textbf{\textit{N}}_4 \hspace{0.2cm} \textbf{\textit{nanosheets.}} \hspace{0.2cm} \textbf{\textit{AFM}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{g-C}}_3\textbf{\textit{N}}_4 \hspace{0.2cm} \textbf{\textit{nanosheets.}} \hspace{0.2cm} \textbf{\textit{AFM}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{g-C}}_3\textbf{\textit{N}}_4 \hspace{0.2cm} \textbf{\textit{nanosheets.}} \hspace{0.2cm} \textbf{\textit{AFM}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{in}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{in}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{in}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{in}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2$ nanosheets, (C) MnO2 nanosheets, and (D) g-C3N4/MnO2. (E) Optimized structure model of g-C3N4/MnO2. (F) Calculated electrostatic potentials for g-C3N4 and the MnO_2 nanosheets, respectively. (G) Charge density difference model of $g-C_3N_4/MnO_2$. Photocatalytic degradation efficiency of (H) RhB and (I) phenol over $g-C_3N_4$. MnO_2 and $g-C_3N_4/MnO_2$ under light irradiation. (J) EPR spectra of DMPO-'OH through irradiation for 120 s in an aqueous dispersion and (K) DMPO-'O²⁻ through irradiation for 120 s in a methanol dispersion in the existence of $g-C_3N_4$, MnO_2 , and $g-C_3N_4/MnO_2$. Mott-Schottky plots for (L) $g-C_3N_4$ and (M) MnO_2 nanosheets. (N) XPS valence band spectra of MnO₂ and g-C₃N₄ nanosheets. (O) Photocatalytic mechanism of $g-C_3N_4/MnO_2$. Reproduced with permission from ref. 137.

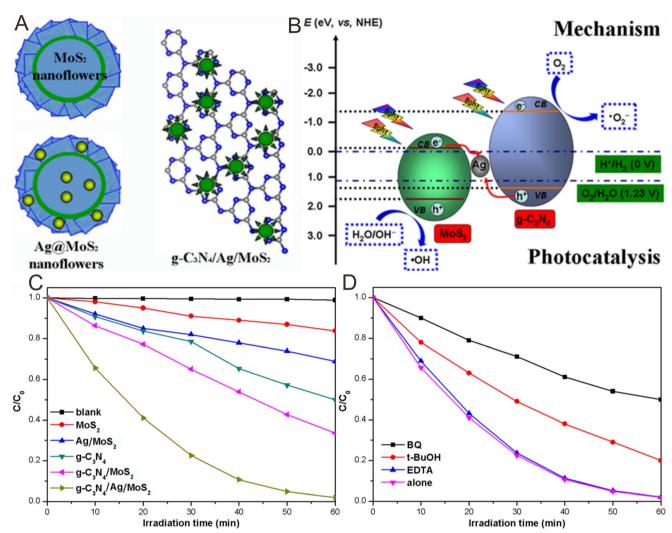


Fig. 13 (A) The nanostructures of Ag/MoS2 and g-C3N4/Ag/MoS2. (B) Diagrammatic sketch for the separation and migration of the photogenerated charges over g-C₃N₄/Ag/MoS₂. (C and D) Photocatalytic property of the fabricated MoS₂, Ag/MoS₂, g-C₃N₄, g-C₃N₄/MoS₂, and g-C₃N₄/Aq/MoS₂ photocatalysts for RhB solution. Reproduced with permission from ref. 247.

significantly enhanced photocatalytic performance, exceeding that of the Ag/MoS2 and g-C3N4/MoS2 systems by 9.43 and 3.56 times, respectively (Fig. 13C and D). The excellent photocatalytic property was mainly because of the synergetic effects of the Ag, g-C₃N₄, and MoS₂ nanophase structures, which led Z-schememechanism-induced rapid separation of the photogenerated pairs and further e^--h^+ enhanced photocatalytic property (Fig. 13B).

5.8. Others

In addition to the above common g-C₃N₄-based Z-scheme photocatalysts, researchers have made many attempts to explore other candidates to fabricate g-C₃N₄-based Z-scheme systems, e.g., g-C₃N₄/ZnO,²⁴⁸ CdS/RGO/g-C₃N₄,²⁴⁹ $g-C_3N_4-RGO-TiO_2$, 250 $MoO_3/g-C_3N_4$, 251 $Cu_3P/g-C_3N_4$, 252 NiTiO₃/g-C₃N₄, ²⁵³ LaFeO₃/g-C₃N₄, ¹²⁰ V₂O₅/g-C₃N₄, ¹⁰⁷ SnO_{2x}/ g-C₃N₄, ²⁵⁴ InVO₄/Bi₂S₃/g-C₃N₄, ²⁵⁵ CDs/g-C₃N₄/SnO₂, ²⁵⁶ gC₃N₄/ZnFe₂O₄/UiO-66,²⁵⁶ g-C₃N₄/BiVO₄/CoFe₂O₄. ²⁵⁷ and They have been extensively utilized as an efficient visiblelight-guided photocatalyst with enhanced photocatalytic degradation activity. The recovery of the photocatalyst is also very important for efficient removal. Thus, the magnetic g-C₃N₄-based Z-scheme photocatalysts^{258,259} are also very useful in this field. The use of g-C₃N₄-based Z-scheme photocatalysts to degrade organic pollutions in actual water bodies is also very important.260 Of course, a more in-depth comprehension of the charge transfer route and photocatalytic mechanism is crucial for further clarification amelioration. and Some theoretical calculations and advanced characterization techniques, such as in situ FTIR, in situ XPS, and synchrotron radiation, are vital for further providing the specific charge transfer route and photocatalytic mechanism. Beyond question, g-C₃N₄-based Z-scheme systems will have enormous potential in photocatalytic organic pollutant degradation.

6. Conclusions and outlook

g-C₃N₄-based photocatalysts have attracted increasing interests because of their easy functionalization, suitable band structure, outstanding physicochemical stability and high photocatalytic property. However, the fast charge recombination and low light utilization greatly limit their practical organic pollution removal application. Luckily, g-C₂N₄-based Z-scheme photocatalysts can greatly boost the spatial separation of the photogenerated carrier and enhance their photocatalytic activity and light utilization. Thus, g-C₃N₄-based Z-scheme photocatalysts exhibit great potential for organic pollution removal. Although many achievements have been obtained, the research studies on g-C₃N₄-based Z-scheme photocatalysts are still at their initial stage, and further explorations are evidently needed. Some opportunities and challenges need to be further emphasized and explored, as shown below:

- (1) A more in-depth comprehension of the charge transfer route and photocatalytic mechanism is crucial for further clarification and amelioration. Some theoretical calculations and characterization techniques, such as in situ FTIR, in situ XPS, and synchrotron radiation, are vital for further providing the specific charge transfer route photocatalytic mechanism. Additionally, the step-scheme (Sscheme) mechanism²⁶¹ is more reasonable and powerful for illustrating the evidently improved photocatalytic property.
- (2) The physicochemical property of the g-C₃N₄-based Z-scheme photocatalysts should be further optimized. The main attempts should concentrate on the construction of advanced semiconductors with high light utilization efficiency, low charge-carrier-transfer resistance, and good physicochemical stability. In addition, engineering (e.g., 0D quantum dots, 1D nanowires and nanorods, 2D nanosheets and 3D sphere and core-shell, etc.) should be considered more.
- (3) The crystalline structure of g-C₃N₄-based Z-scheme photocatalysts is also vital for the enhanced charge migration efficiency. Normally, the transfer efficiency of photogenerated charges will be greatly decreased owing to the defects, mismatch parameters, configurations. Therefore, the optimization of the crystalline structure of the g-C₃N₄-based Z-scheme photocatalysts is a promising method for improving their photocatalytic performance.
- (4) The photocatalytic performance of g-C₃N₄-based Z-scheme systems can be further improved via fabricating ternary or multicomponent systems. A practical method is to load suitable co-catalysts on the g-C₃N₄-based Z-scheme photocatalysts that will be very good for further enhancing its e⁻-h⁺ separation and light utilization.
- (5) Considering the unique advantages of g-C₃N₄-based Z-scheme systems, widening their application scope (i.e., conversion, desulfurization, photosynthesis) is critical to building a sustainable society.
- (6) In regard to g-C₃N₄-based Z-scheme systems for organic pollution removal, only considering

photocatalytic property is not enough. Other aspects, including cost, environment, and ecology, should also be focused on.

(7) In order to achieving a practical application of g-C₃N₄based Z-scheme systems, some efficient enhancement approaches, e.g., morphology regulation, loading co-catalysts, interface engineering, doping and vacancy, and exposing more reactive facets, should be highly considered.

Looking to the future, there has been a lot of opportunities and challenges for g-C₃N₄-based Z-scheme photocatalysts for organic pollution removal. It is believed that this review can provide some new and advanced insights for guiding the rational design of g-C₃N₄-based Z-scheme systems for organic pollution removal. Certainly, the g-C₃N₄based Z-scheme systems can achieve practical industrial applications in the near future.

Conflicts of interest

There are no conflicts of interest to declare.

Acknowledgements

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