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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 g-C₃N₄-based Z-scheme photocatalysts for organic pollution removal, beginning with the fundamentals of g-C₃N₄-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.

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,

such as adsorption,^{7–10} electrocatalysis,^{11,12} and 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, suitable electronic and band structure, excellent 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.



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To enhance the photocatalytic property of g-C₃N₄, various approaches have been proposed, *e.g.*, doping,^{52,53} obtaining porous architectures,^{54,55} achieving highly crystalline structures,⁵⁶ metal loading,^{57,58} 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.



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Although there have been many inspired reviews on g-C₃N₄-based photocatalysts 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-C₃N₄-based 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-C₃N₄-based Z-scheme photocatalysts?

Before introducing the g-C₃N₄-based Z-scheme 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/flowering 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 e⁻ in 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.⁷⁴ 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-C₃N₄-based Z-scheme systems, including g-C₃N₄-based direct Z-scheme (Fig. 1B) without an electron mediator and g-C₃N₄-based semiconductor-conductor-semiconductor (SCS) (Fig. 1C).



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.^{106,107} 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/g-C₃N₄,¹¹⁶ 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 favorable migration of photogenerated carriers.¹¹⁸ Moreover, the p-n Z-scheme systems (*e.g.*, C₃N₄-Pd-Cu₂O,¹¹⁹ LaFeO₃/g-C₃N₄,¹²⁰ g-C₃N₄/CaFe₂O₄,¹²¹ black 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.*, self-confirmation *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.

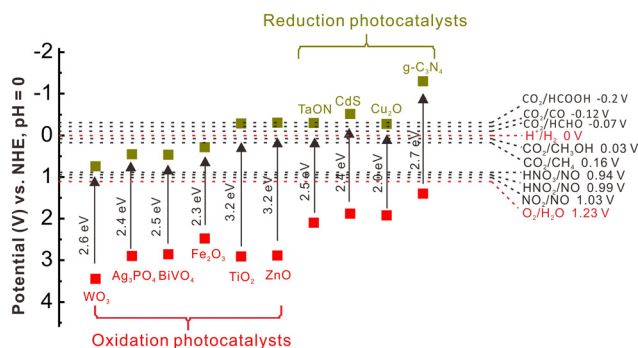


Fig. 2 Standard redox potentials of some commonly used semiconductors. Reproduced with permission from ref. 35.

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 by 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 display enhanced photocatalytic performance. This 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-workers¹²⁸ proved the successful fabrication of the g-C₃N₄/CoTiO₃ direct Z-scheme heterojunctions through analyzing the 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

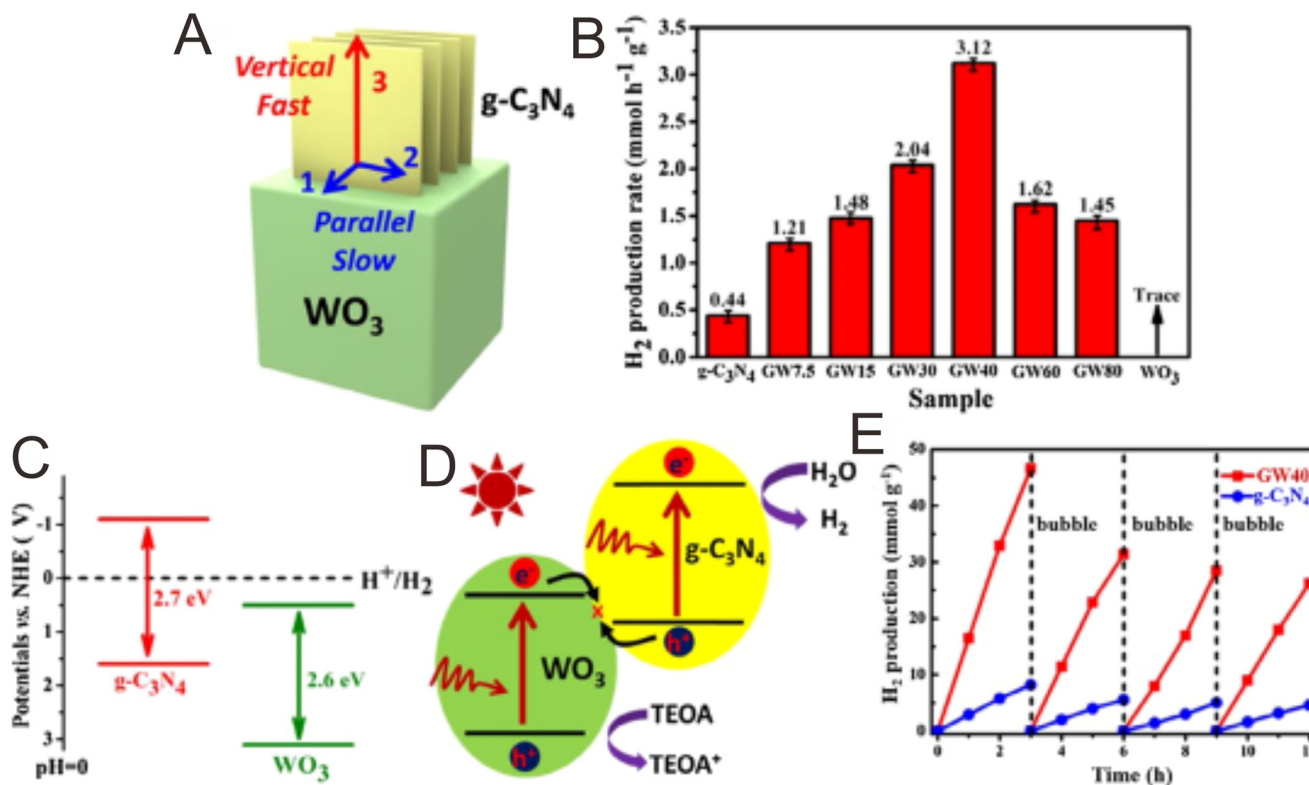


Fig. 3 (A) The growth and extension of g-C₃N₄ parallel with and perpendicular to the flat surface of the WO₃ nanocuboids. (B) Comparison of the photocatalytic H₂ production properties. (C) Band structure of g-C₃N₄ and WO₃. (D) The direct Z-scheme charge migration path in the g-C₃N₄/WO₃ composites. (E) 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.

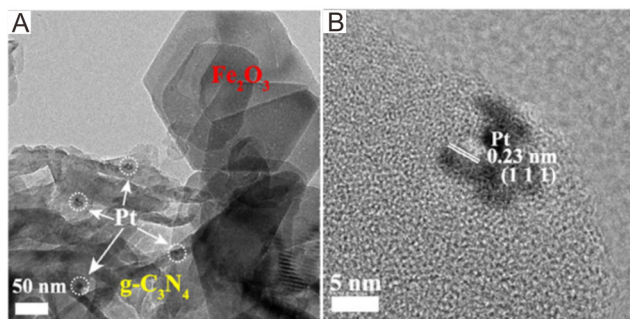


Fig. 4 (A) TEM image and (B) HRTEM image of the Fe₂O₃/g-C₃N₄ composites after photodeposition of Pt nanoparticles. Reproduced with permission from ref. 130.

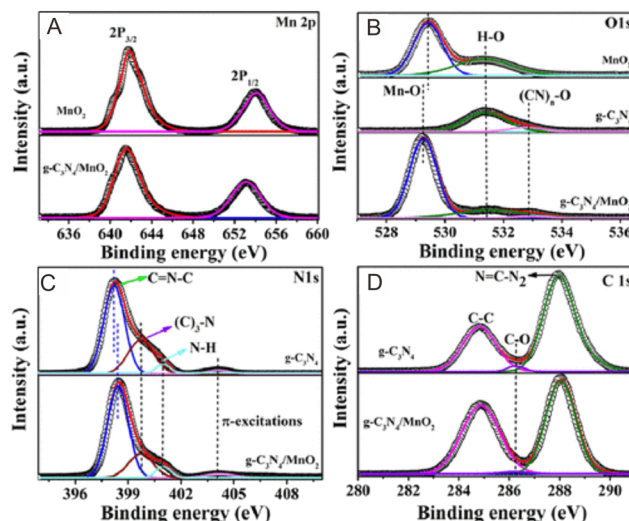


Fig. 5 XPS spectra of (A) Mn 2p, (B) O 1s, (C) N 1s, and (D) C 1s for g-C₃N₄, MnO₂, and g-C₃N₄/MnO₂. Reproduced with permission from ref. 137.

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 $g\text{-C}_3\text{N}_4/\text{MnO}_2$,¹³⁷ the Mn 2p spectrum of the MnO_2 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 $g\text{-C}_3\text{N}_4$ sample only exhibited the adsorbed oxygen species and other oxygen impurities (Fig. 5B). In view of $g\text{-C}_3\text{N}_4/\text{MnO}_2$, 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 $g\text{-C}_3\text{N}_4$ (Fig. 5C) can be divided into three main peaks at 398.2, 399.7, and 401.2 eV, which can be assigned to the sp^2 -hybridized nitrogen (C=N-C), bridging nitrogen ((C)₃-N), and N-H bonding in the $g\text{-C}_3\text{N}_4$ framework, respectively. When compared with the MnO_2 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 MnO_2 onto the $g\text{-C}_3\text{N}_4$ nanosheets with electrons flowing from $g\text{-C}_3\text{N}_4$ to MnO_2 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 SPV techniques to study the carrier transfer path of Z-scheme systems is an efficient technology. Guan and co-workers¹⁴⁴ reported a $g\text{-C}_3\text{N}_4$ -based Z-scheme system through encapsulation of $g\text{-C}_3\text{N}_4$ into hollow In_2O_3 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 of 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 $g\text{-C}_3\text{N}_4$ to the interior of In_2O_3 , further illustrating the Z-scheme mechanism. Therefore, the SPV strategy is also very promising for the confirmation of the Z-scheme mechanism.

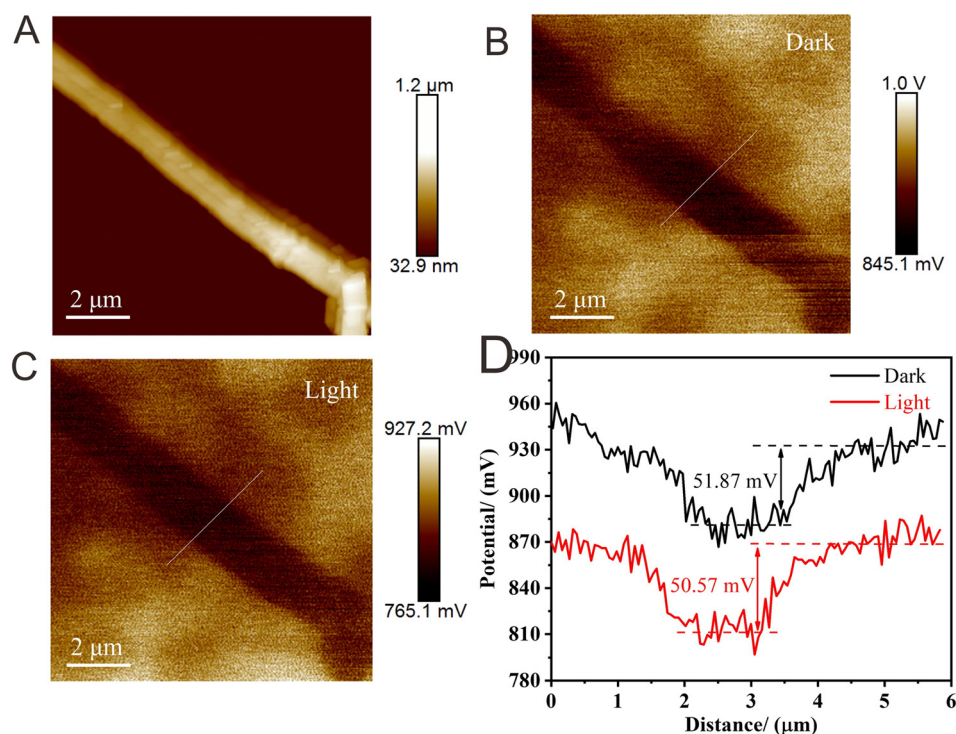


Fig. 6 (A) Atomic force microscopy image of the samples. (B) KPFM potential image of the samples under dark conditions. (C) KPFM potential image of the samples under light illumination. (D) The corresponding surface curves correspond to the white light. Reproduced with permission from ref. 144.

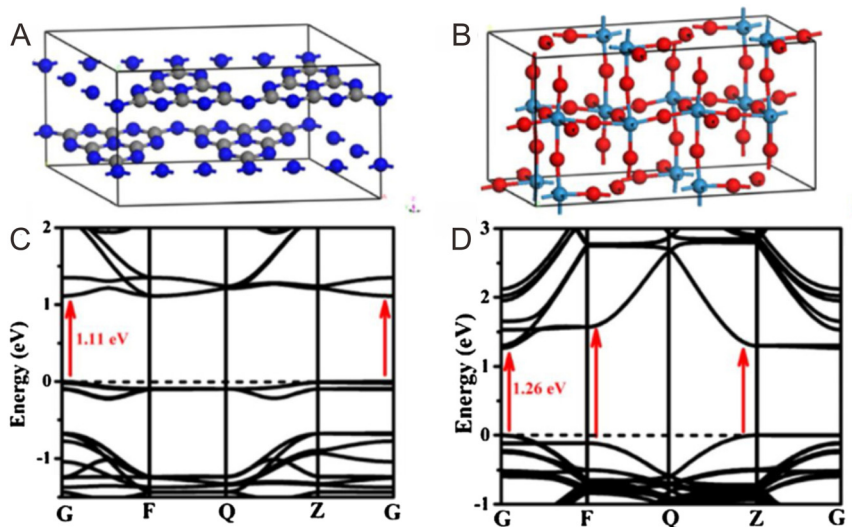


Fig. 7 Crystal structures of (A) g-C₃N₄ and (B) monoclinic WO₃. The calculated electronic band structures of (C) g-C₃N₄ and (D) WO₃. Reproduced with permission from ref. 127.

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):

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

$$\nu = \hbar k/m^* \quad (2)$$

$$D = m_h^*/m_e^* \quad (3)$$

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 second-order 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	G-F direction	G-Z direction	F-Q direction	Z-Q direction
g-C ₃ N ₄	m_h^*	0.72	71	—	—
	m_e^*	0.53	1.8	—	—
	D	1.36	39.44	—	—
WO ₃	m_h^*	0.53	—	0.49	0.51
	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-C₃N₄ ($D = 1.36$) 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 g-C₃N₄-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 for 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, and 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} The 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 of g-C₃N₄

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-workers¹⁷⁰ prepared a TiO₂/g-C₃N₄/RGO ternary Z-scheme heterojunction *via* an electrospinning coupled approach. The crystal structure, morphology, composition, specific surface area,

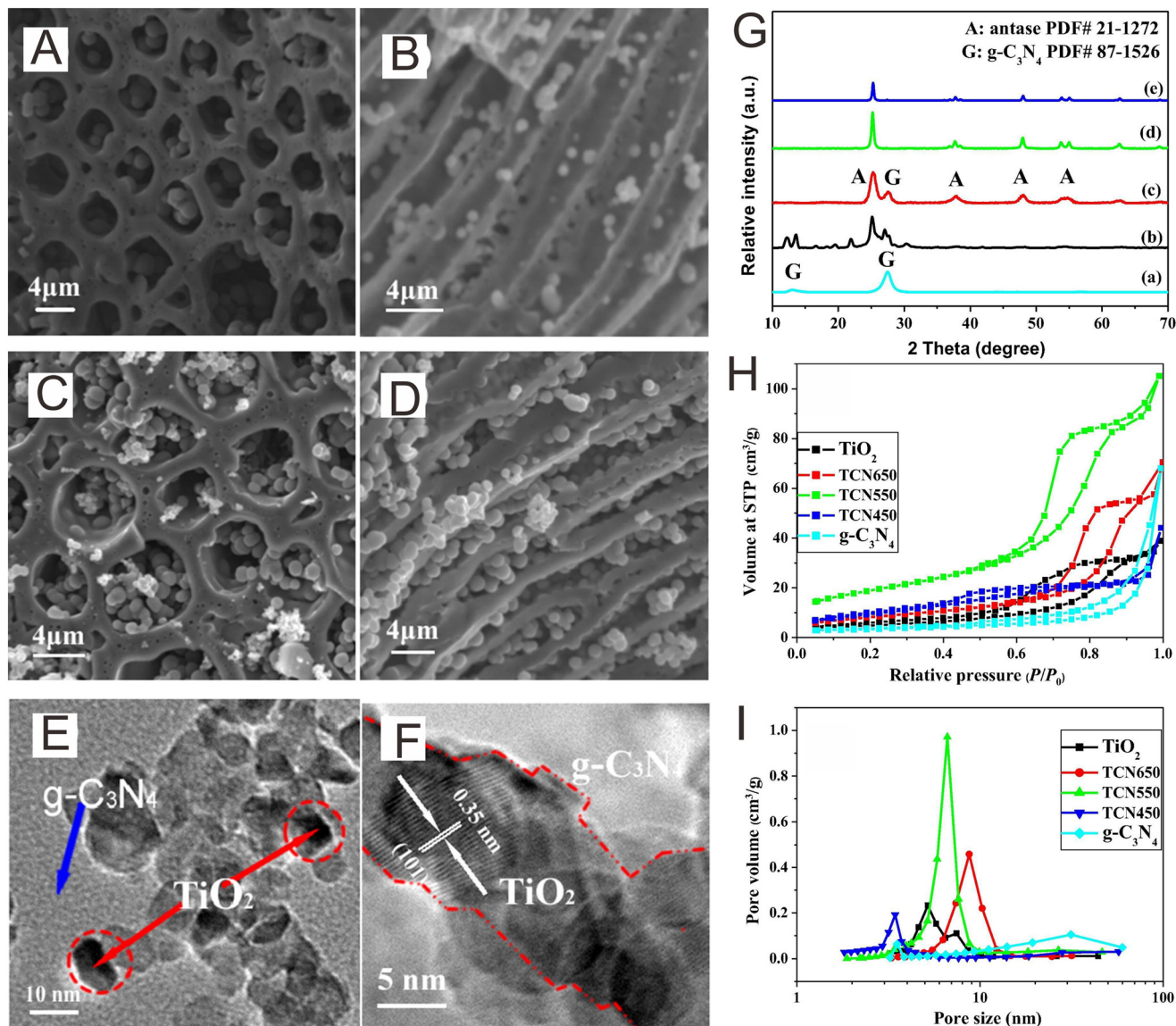


Fig. 8 SEM images at different perspective views of (A and B) the pure hierarchical TiO₂ framework and (C and D) the TiO₂/g-C₃N₄ Z-scheme photocatalyst calcination at 550 °C (denoted as TCN550). TEM (E) and HRTEM (F) images of the TCN550. XRD (G), N₂ adsorption isotherms (H), and pore size distribution (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 self-assembly approach for preparing g-C₃N₄/ZnO Z-scheme photocatalysts. In this system, the electrostatic force between g-C₃N₄ 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.¹⁷⁶ Therefore, the self-assembly 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

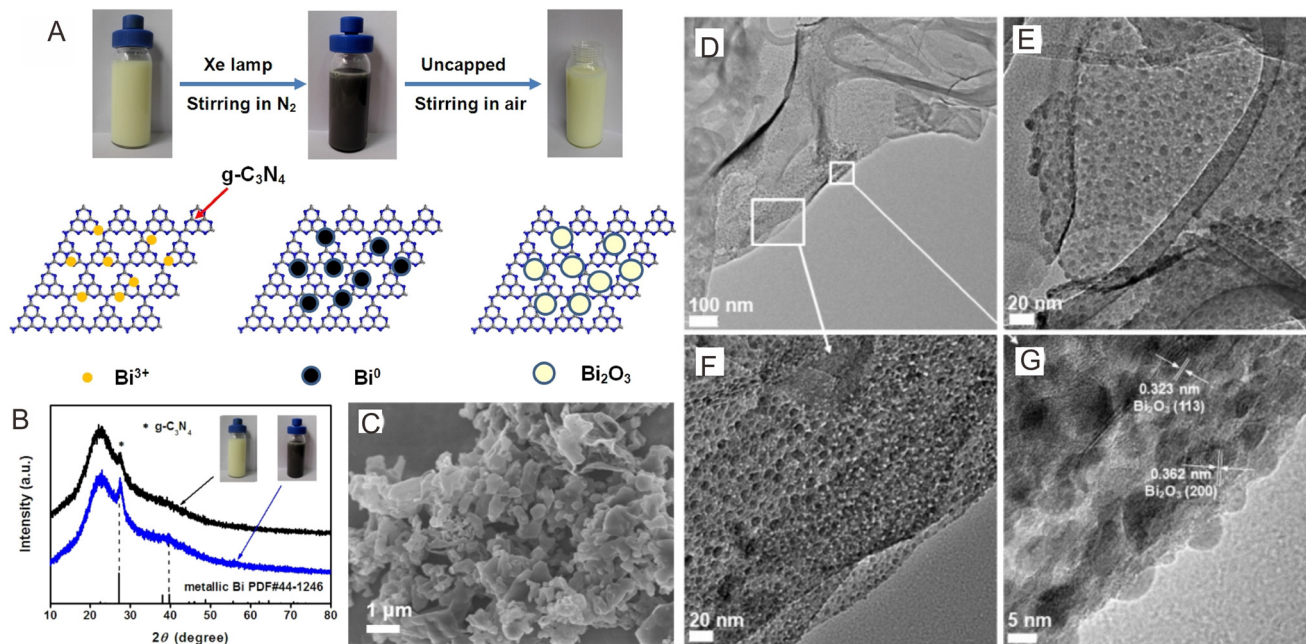


Fig. 9 (A) Diagrammatic sketch of the preparation of $\text{Bi}_2\text{O}_3/\text{g-C}_3\text{N}_4$. (B) XRD patterns of the intermediate products. (C) FESEM image of $\text{Bi}_2\text{O}_3/\text{g-C}_3\text{N}_4$. TEM (D–F) and HRTEM (G) images of $\text{Bi}_2\text{O}_3/\text{g-C}_3\text{N}_4$. Reproduced with permission from ref. 180.

promising for preparing Z-scheme photocatalysts. A $\text{Bi}_2\text{O}_3/\text{g-C}_3\text{N}_4$ direct Z-scheme photocatalyst was fabricated through the simple photoreductive deposition of Bi^{3+} on the $\text{g-C}_3\text{N}_4$ surface under light illumination, followed by air oxidation of Bi^{3+} to Bi_2O_3 (Fig. 9A).¹⁸⁰ The uniform dispersion of quantum-sized Bi_2O_3 on the $\text{g-C}_3\text{N}_4$ nanosheet surface could be observed from the XRD pattern, FESEM, TEM and HRTEM images (Fig. 9B–G). Wu and co-workers¹⁸¹ reported a novel $\text{BiVO}_4(040)\text{-Ag@Cds}$ Z-scheme photocatalyst in which Ag was selectively photo-deposited on the (040) facets of BiVO_4 , and the Cds nanorods were decorated on $\text{BiVO}_4(040)\text{-Ag}$. Thus, the photo-deposition strategy is also a promising technology for 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 $\text{g-C}_3\text{N}_4/\text{WO}_3$ Z-scheme photocatalysts *via* grinding, impregnation assisted with sonication, and planetary ball-milling methods. Different from the agglomerated WO_3 decorated on the $\text{g-C}_3\text{N}_4$ nanoplate prepared through the first two methods, WO_3 prepared through planetary ball-milling strategy was uniformly dispersed on the $\text{g-C}_3\text{N}_4$

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

Among the various fabrication strategies, deposition-precipitation, 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 weak 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 (BB), 2,4-dichlorophenol (2,4-DCP), tetracycline (TC), sulfamethoxazole (SMX), indomethacin (IDM), and others

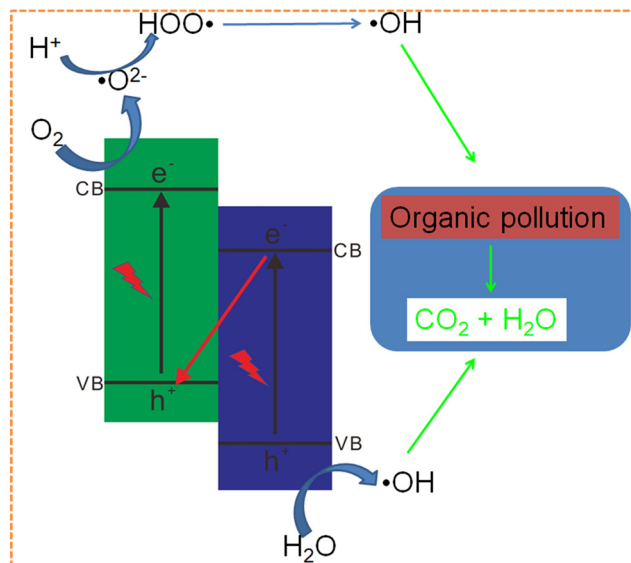


Fig. 10 Diagrammatic explanation of organic pollutant degradation over $g\text{-C}_3\text{N}_4$ -based Z-scheme photocatalysts.

have recently been excessively discharged to soil and water, which have negative influences on the environment and human health.^{193–201} $g\text{-C}_3\text{N}_4$ -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. $g\text{-C}_3\text{N}_4/\text{TiO}_2$ Z-scheme photocatalysts

TiO_2 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 TiO_2 with $g\text{-C}_3\text{N}_4$ 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 photocatalytic properties by $g\text{-C}_3\text{N}_4/\text{TiO}_2$ Z-scheme photocatalysts. Dong and co-workers²⁰⁵ fabricated an all-solid-state (ASS) Z-scheme photocatalyst through decorating $g\text{-C}_3\text{N}_4$ nanosheets on TiO_2 nanotube arrays (TNTAs). The as-prepared photocatalyst displayed improved photocatalytic property for RhB degradation, which was about 4 times greater than that exhibited by pure TiO_2 TNTAs. Ho and co-workers²⁰⁶ prepared the $g\text{-C}_3\text{N}_4$ quantum dots (QDs)-modified rutile TiO_2 ($r\text{TiO}_2$) ($g\text{-C}_3\text{N}_4$ QDs- $r\text{TiO}_2$) hybrid *via* calcination of the mixture of P25 TiO_2 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 BiVO_4 have been demonstrated to be the active reduction and oxidation sites, respectively.²⁰⁷ Raj and co-workers²⁰⁸ fabricated a $g\text{-C}_3\text{N}_4/\text{TiO}_2/\text{CuO}$ 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 TiO_2 on the photocatalytic activity. They first fabricated TiO_2 nanosheets (NSs) and TiO_2 nanoboxes (NBs), and then combined them with $g\text{-C}_3\text{N}_4$ to prepare TiO_2 NS/ $g\text{-C}_3\text{N}_4$ and TiO_2 NBs/ $g\text{-C}_3\text{N}_4$, respectively (Fig. 11A and B). The $\{001\}$ facets of TiO_2 NSs came into contact with $g\text{-C}_3\text{N}_4$ (Fig. 11A and C), and the $\{101\}$ facets of TiO_2 NBs came into contact with $g\text{-C}_3\text{N}_4$ (Fig. 11B and D). Under visible light illumination, the photogenerated carriers on TiO_2 were separately accumulated on the $\{101\}$ and $\{001\}$ facets.²¹⁰ The photogenerated electrons were subsequently centered on the $\{101\}$ facets of the TiO_2 NBs, which could transfer to $g\text{-C}_3\text{N}_4$ for photocatalytic reactions, which is in line with the direct Z-scheme mechanism (Fig. 11F). Thus, the e^- - h^+ separation efficiency of TiO_2 NBs/ $g\text{-C}_3\text{N}_4$ is much higher than that of TiO_2 NSs/ $g\text{-C}_3\text{N}_4$, and the TiO_2 NBs/ $g\text{-C}_3\text{N}_4$ 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. $g\text{-C}_3\text{N}_4/\text{WO}_3$ Z-scheme photocatalysts

WO_3 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 TiO_2 , WO_3 possesses a smaller band gap (2.7 eV) and more positive CB level. Thus, it is also favorable for combining with $g\text{-C}_3\text{N}_4$ to construct Z-scheme photocatalysts. The Chen group²¹³ developed a $\text{WO}_3/g\text{-C}_3\text{N}_4$ Z-scheme photocatalyst. The reaction rate constants for degrading MB and BF over $\text{WO}_3(5.0 \text{ wt}\%)/g\text{-C}_3\text{N}_4$ were found to be 4.77 and 3.84 times greater than that of the bare $g\text{-C}_3\text{N}_4$, respectively. The Jia group²¹⁴ developed a new bifunctional $\text{WO}_3/g\text{-C}_3\text{N}_4$ Z-scheme photocatalyst by *in situ* liquid phase strategy. The photocatalytic degradation efficiency of the 30 wt% $\text{WO}_3/g\text{-C}_3\text{N}_4$ was found to be 2.5 and 2.7 times higher than the individual $g\text{-C}_3\text{N}_4$ and WO_3 , respectively. Kang and co-workers²¹⁵ fabricated $\text{WO}_3/g\text{-C}_3\text{N}_4$ 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 $\text{WO}_3/g\text{-C}_3\text{N}_4$ 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 $g\text{-C}_3\text{N}_4/\text{WO}_3$ Z-scheme photocatalyst through the hydrothermal strategy. Benefiting from high specific areas, C-substitution, and Z-scheme mechanism, the

Table 3 Comparison of the photocatalytic organic pollution degradation performance over g-C₃N₄-based Z-scheme photocatalysts

Photocatalysts	Preparation strategy	Mass (mg)	Light source	Target pollutant/initial concentration	Degradation time (min) and rate (%)	K_{app}^a [10^{-2} min ⁻¹]	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 mg L ⁻¹	120 (~97)	0.95	208
g-C ₃ N ₄ /TiO ₂ -HNB	Microwave-assisted strategy	—	3 W LED lamp, 365 nm	Brilliant red X-3B/1.0 × 10 ⁻⁴ mmol L ⁻¹	45 (~99)	5.1	209
WO ₃ /g-C ₃ N ₄	Ball milling and heat treatment methods	10	500 W xenon lamp, >470 nm	MB and BF/0.9 × 10 ⁻⁵ and 1.0 × 10 ⁻⁵ mol L ⁻¹	MB 120 (~99) BF 120 (~97)	—	213
WO ₃ /g-C ₃ N ₄	<i>In situ</i> liquid phase process	50	300 W xenon lamp, >400 nm	MB/50 mg L ⁻¹	90 (~97)	1.90	214
WO ₃ /g-C ₃ N 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 strategy	100	500 W xenon lamp, >420 nm	TC/10 mg L ⁻¹	60 (~70)	—	217
WO ₃ /g-C ₃ N ₄ /Bi ₂ O ₃	One step co-calcination way	100	300 W xenon lamp, >420 nm	TC/10 mg L ⁻¹	60 (~80)	2.37	159
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 ₂ O ₃ /g-C ₃ N ₄	Ball milling and heat treatment methods	5	500 W xenon lamp, 470 nm	MB and RhB/1.1 × 10 ⁻⁵ and 1.0 × 10 ⁻⁵ mol L ⁻¹	MB 120 (~76) RhB 120 (~73)	2.53 and 1.01	223
g-C ₃ N ₄ /Bi ₂ O ₃ /BiPO ₄	Calcination route	15	—	MO/10 mg L ⁻¹	160 (~88)	1.15	225
β-Bi ₂ O ₃ /g-C ₃ N ₄	Hydrothermal calcination approach	40	350 W xenon lamp, >420 nm	RhB/1 × 10 ⁻⁵ mol/L	80 (~97)	4.22	226
g-C ₃ N ₄ /Bi ₄ O 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 method	50	300 W xenon lamp, 350–780 nm	2,4-DCP/20 mg L ⁻¹	120 (>99)	1.13	228
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 lamp, >420 nm	TCP/20–100 mg L ⁻¹	120 (>99)	3.15	229
Ag-decorated g-C ₃ N ₄ /Bi ₂ WO ₆	Polymerization method	40	300 W xenon lamp, 400 nm	RhB/10 mg L ⁻¹	90 (>99)	—	230
Bi ₃ O ₄ Cl/g-C ₃ N ₄	Solid-phase calcination method	50	250 W xenon lamp, >420 nm	RhB and TC/10 and 10 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 lamp, 420 nm	RhB and TC/10 and 10 mg L ⁻¹	RhB 7.5 (>99) TC 60 (>97)	— and —	232
Bi ₁₂ GeO ₂₀ /g-C ₃ N ₄	Sol-gel method	300	300 W Dy lamp, 400 nm	RhB/5 mg L ⁻¹	50 (>99)	2.55	233
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 ₃ PO ₄ /g-C ₃ N ₄	<i>In situ</i> precipitation method	5	300 W xenon lamp, >400 nm	SMX/1 mg L ⁻¹	10 (~98)	6.3	238
Ag ₃ PO ₄ /g-C ₃ N ₄	<i>In situ</i> precipitation method	30	300 W xenon lamp	MO/10 mg L ⁻¹	5 (>99)	—	239
g-C ₃ N ₄ /Ag ₂ WO	<i>In situ</i> precipitation method	30	300 W xenon lamp	MO/10 mg L ⁻¹	150 (~98)	—	240
Ag ₂ CrO ₄ /g-C ₃ N 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 method	100	300 W xenon lamp	MO and MB/10 and 10 mg L ⁻¹	MO 10 (~97) RhB 10 (~96)	— and —	117
Ag ₂ CrO ₄ /Ag/g-C ₃ N ₄	<i>In situ</i> growth and photoreduction methods	50	500 W xenon lamp, <420 nm	2,4-DCP/10 mg L ⁻¹	120 (~96)	1.52	243
g-C ₃ N ₄ /Ag/MoS ₂	<i>In situ</i> growth and precipitation methods	100	300 W xenon lamp, >420 nm	RhB/20 mg L ⁻¹	60 (>99)	—	247
g-C ₃ N ₄ /ZnO	Ultrasonically exfoliated method	100	300 W xenon lamp, >420 nm	4-CP/10 ⁻⁴ mol L ⁻¹	90 (>99)	5.09	248
CdS/RGO/g-C ₃ N ₄	Hydrothermal method	20	350 W xenon lamp	Atrazine/—	300 (~92)	0.71	249
g-C ₃ N ₄ -RGO-TiO ₂	Liquid-precipitation strategy	50	300 W xenon lamp, >420 nm	RhB/30 mg L ⁻¹	180 (~92)	1.37	250
MoO ₃ /g-C ₃ N ₄	Calcination method	30	350 W xenon lamp, >420 nm	TC/20 mg L ⁻¹	90 (~90)	2.23	251
p-type Cu ₃ P/n-type g-C ₃ N ₄	Calcination method	10	300 W xenon lamp, 400 nm	RhB/—	20 (>99)	—	252

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 ₂ O ₅ /g-C ₃ N ₄	<i>In situ</i> growth strategy	50	250 W xenon lamp, 420 nm	RhB and TC/10 and 10 mg L ⁻¹	120 (~77)	4.91 and —	107
SnO ₂ /g-C ₃ N ₄	Calcination method	100	350 W xenon lamp, >420 nm	RhB/10 mg L ⁻¹	40 (>99)	1	254
InVO ₄ /Bi ₂ S ₃ /g-C ₃ N ₄	Wet-impregnation method	200	300 W xenon lamp, >420 nm	RhB19/15 mg L ⁻¹	100 (~96)	0.57	255
CDs/g-C ₃ N ₄ /SnO ₂	Thermal polymerization	10	300 W xenon lamp, >420 nm	IDM/10 mg L ⁻¹	40 (>99)	2.33	256
g-C ₃ N ₄ /ZnFe ₂ O ₄ /UiO-66	Hydrothermal method	—	300 W xenon lamp, >420 nm	RhB/—	40 (>99)	4.89	256
g-C ₃ N ₄ /BiVO ₄ /CoFe ₂ O ₄	High-temperature solid-state route	50	300 W xenon lamp, >400 nm	RhB/—	25 (>99)	2.84	257
g-C ₃ N ₄ /g-C ₃ N 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 band-gap, 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).^{219,220} 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.*, Bi₂O₃,^{159,180,223–225} β-Bi₂O₃,²²⁶ Bi₄O₇,²²⁷ Bi₂WO₆,^{156,228–230} Bi₃O₄Cl,²³¹ BiOBr,²³² Bi₁₂GeO₂₀,²³³ Bi₂MoO₆,²³⁴ 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, and highly efficient photocatalytic performance.^{235–237} For example, Chen and co-workers²²³ 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 than 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

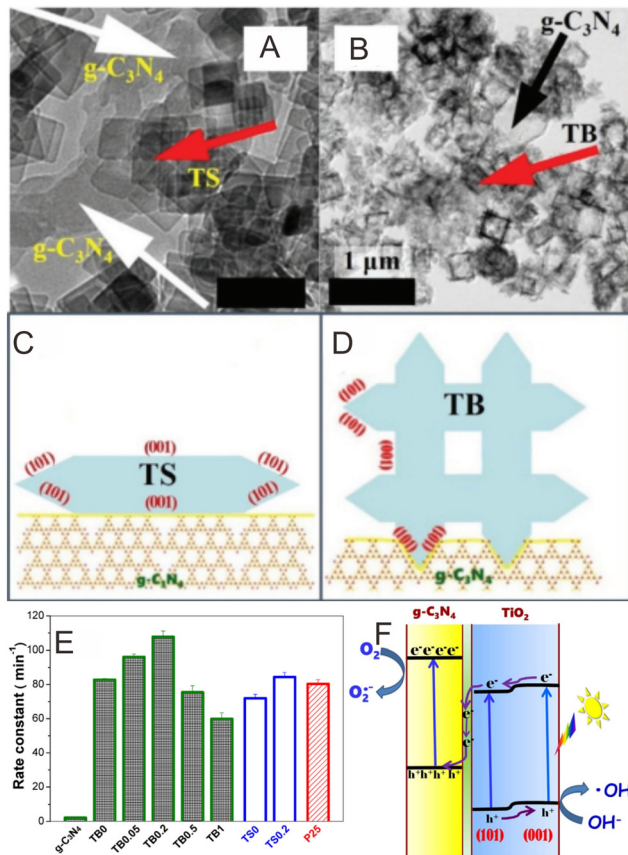


Fig. 11 TEM images of (A) TiO₂ NSs/g-C₃N₄ and (B) TiO₂ NBs/g-C₃N₄. Diagrammatic sketch of the contact interface between TiO₂ and g-C₃N₄ on (C) TiO₂ NSs/g-C₃N₄ and (D) TiO₂ NBs/g-C₃N₄. (E) Comparison of the photocatalytic activity of the synthesized photocatalysts for the photocatalytic degradation of 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 as-prepared 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 Bi₁₂GeO₂₀/g-C₃N₄ 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 g-

C₃N₄ to fabricate Z-scheme photocatalysts for enhancing photocatalytic properties owing to their suitable CB and CB values.

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 Ag-containing semiconductors/g-C₃N₄ SCS Z-scheme photocatalysts can achieve more effective photocatalytic performance.²⁴² The Guo group¹¹⁷ fabricated various Ag@AgBr/g-C₃N₄ Z-scheme photocatalysts *via* the 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 co-workers²⁴³ 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-C₃N₄/g-C₃N₄ Z-scheme photocatalysts showed better 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 ~0.4–0.6 μm (Fig. 13A). The prepared g-C₃N₄/Ag/MoS₂ showed

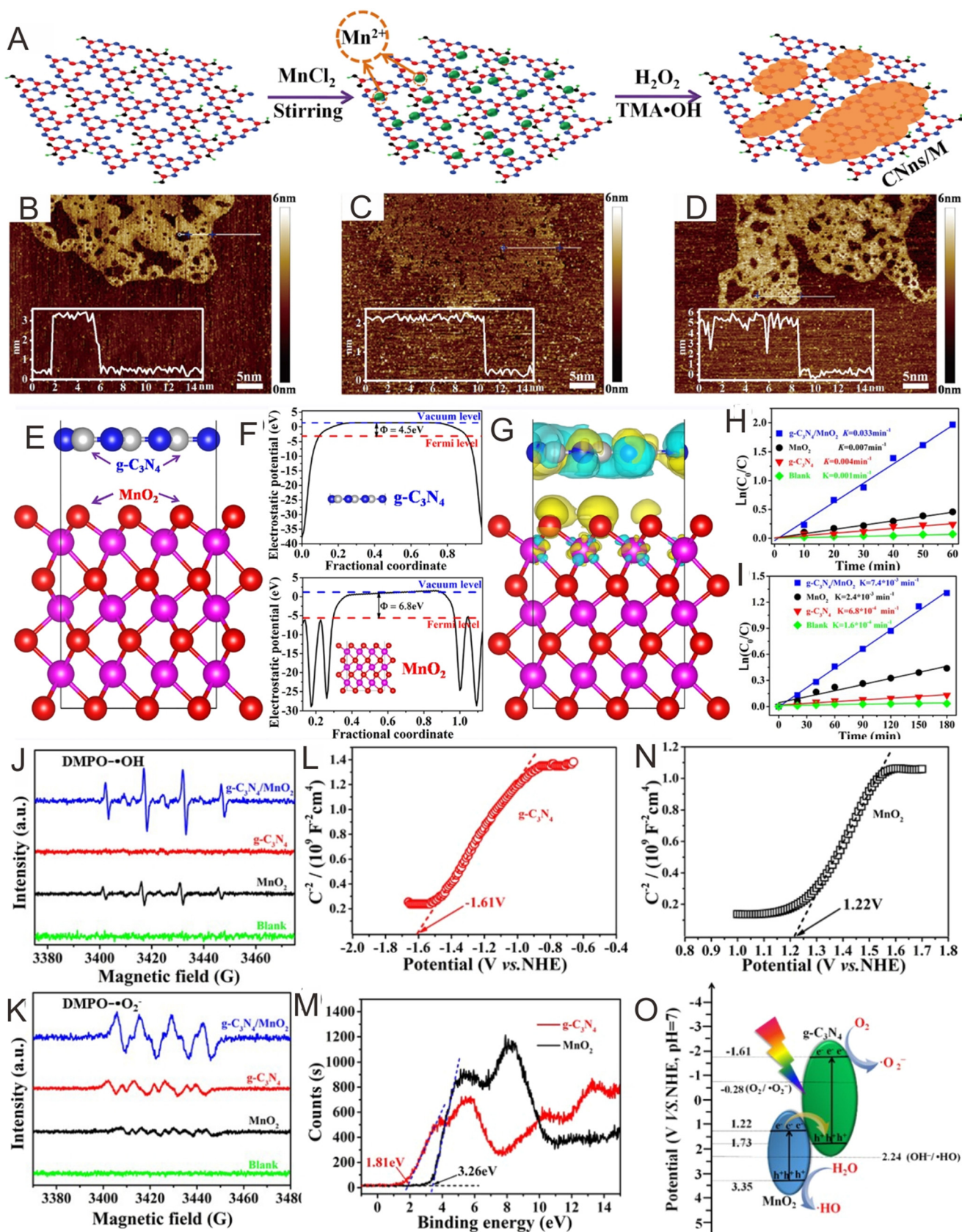


Fig. 12 (A) Diagrammatic sketch of $g\text{-C}_3\text{N}_4/\text{MnO}_2$ prepared through *in situ* growth of MnO_2 on the $g\text{-C}_3\text{N}_4$ nanosheets. AFM images of the (B) $g\text{-C}_3\text{N}_4$ nanosheets, (C) MnO_2 nanosheets, and (D) $g\text{-C}_3\text{N}_4/\text{MnO}_2$. (E) Optimized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

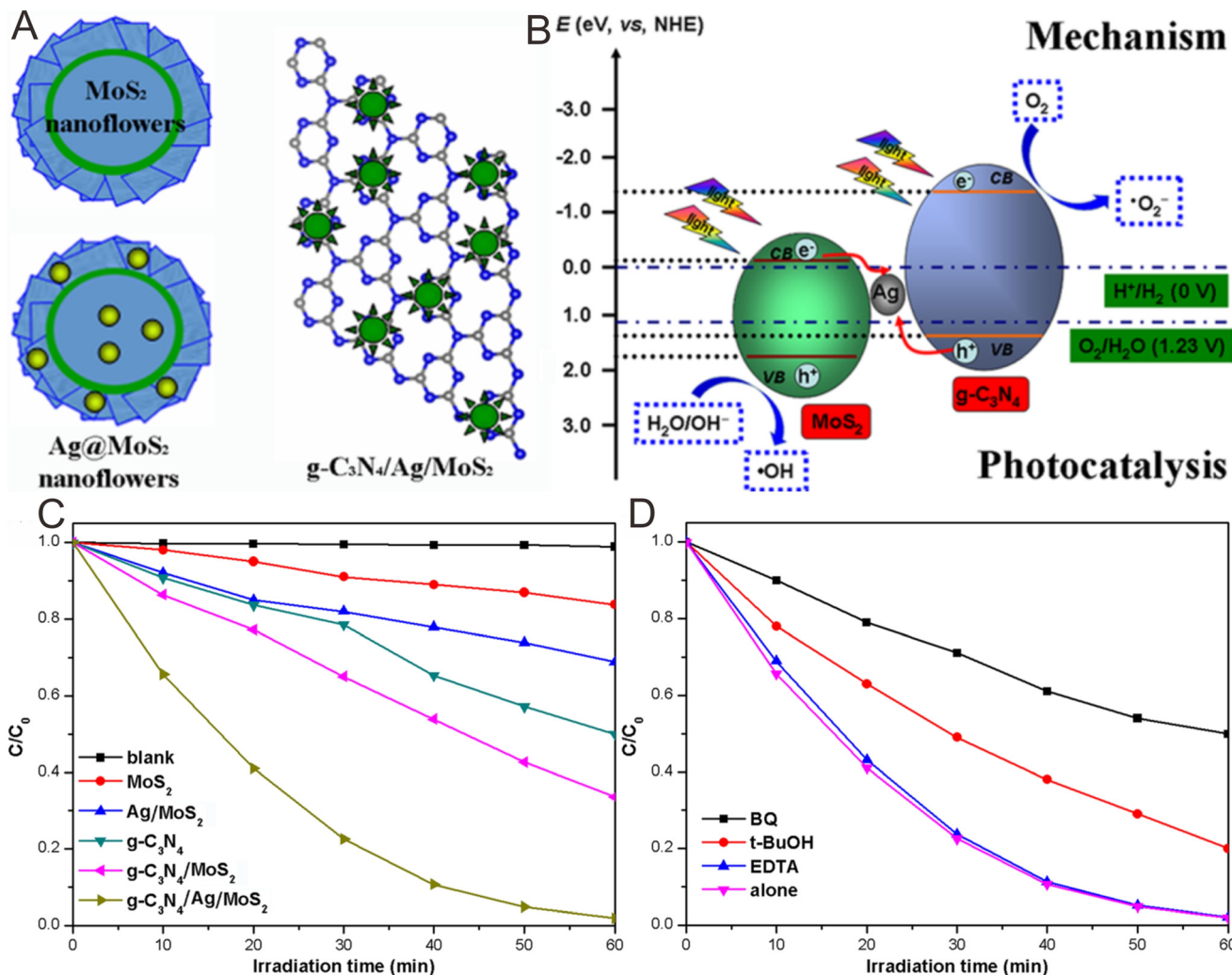


Fig. 13 (A) The nanostructures of Ag/MoS₂ and g-C₃N₄/Ag/MoS₂. (B) Diagrammatic sketch for the separation and migration of the photogenerated charges over g-C₃N₄/Ag/MoS₂. (C and D) Photocatalytic property of the fabricated MoS₂, Ag/MoS₂, g-C₃N₄, g-C₃N₄/MoS₂, and g-C₃N₄/Ag/MoS₂ photocatalysts for RhB solution. 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 synergistic effects of the Ag, g-C₃N₄, and MoS₂ nanophase structures, which led to a Z-scheme-mechanism-induced rapid separation of the photogenerated e⁻-h⁺ pairs and further 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-C₃N₄-RGO-TiO₂,²⁵⁰ MoO₃/g-C₃N₄,²⁵¹ Cu₃P/g-C₃N₄,²⁵² 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 g-

C₃N₄/ZnFe₂O₄/UiO-66,²⁵⁶ and g-C₃N₄/BiVO₄/CoFe₂O₄.²⁵⁷ They have been extensively utilized as an efficient visible-light-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 and amelioration. 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-C₃N₄-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 and photocatalytic mechanism. Additionally, the step-scheme (S-scheme) 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, dimension 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 of lattice parameters, and electronic 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.*, energy gas conversion, desulfurization, organic photosynthesis) is critical to building a sustainable society.

(6) In regard to g-C₃N₄-based Z-scheme systems for organic pollution removal, only considering the

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-C₃N₄-based 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-C₃N₄-based 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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