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# MINI REVIEW

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# Recent advances on g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts for organic pollutant removal

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Graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>)-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-C<sub>3</sub>N<sub>4</sub>based 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-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts are discussed. Various typical g-C<sub>3</sub>N<sub>4</sub>-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.<sup>1-6</sup> 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-C<sub>3</sub>N<sub>4</sub>)-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-C<sub>3</sub>N<sub>4</sub> is still far from practical



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production demand owing to the fast electron-hole (e<sup>-</sup>-h<sup>+</sup>) recombination and poor light absorption capacity. 48-51 Hence, it is crucial to explore new g-C<sub>3</sub>N<sub>4</sub>-based photocatalysts with strong light absorption capacity and high e<sup>-</sup>-h<sup>+</sup> separation for organic pollutant removal.



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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.<sup>72</sup> To overcome this disadvantage, an emerging g-C<sub>2</sub>N<sub>4</sub>-based Z-scheme system was proposed by the Yu group<sup>73</sup> 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, <sup>76</sup> g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts have achieved increasing attention for organic pollutant removal.

To enhance the photocatalytic property of g-C<sub>3</sub>N<sub>4</sub>, various approaches have been proposed, e.g., doping, 52,53 obtaining



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Although there have been many inspired reviews on gphotocatalysts C<sub>3</sub>N<sub>4</sub>-based for various applications, 32,33,71,77-88 a comprehensive review about g-C<sub>3</sub>N<sub>4</sub>-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-C<sub>3</sub>N<sub>4</sub>based Z-scheme photocatalysts are firstly discussed. Subsequently, the characterization strategies and preparation methods for g-C<sub>3</sub>N<sub>4</sub>-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-C<sub>3</sub>N<sub>4</sub>-based Before introducing the photocatalysts, it is crucial to fully comprehend g-C<sub>3</sub>N<sub>4</sub>-based type-II heterojunctions, e.g., CeO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub>, <sup>89</sup> Co<sub>3</sub>O<sub>4</sub>/g-C<sub>3</sub>N<sub>4</sub>, <sup>90</sup> Cu<sub>2</sub>O/g-C<sub>3</sub>N<sub>4</sub>, <sup>91</sup> Fe<sub>2</sub>O<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub>, <sup>92</sup> g-C<sub>3</sub>N<sub>4</sub>/g-C<sub>3</sub>N<sub>4</sub>, <sup>93</sup> h-BN/flowerring g-C<sub>3</sub>N<sub>4</sub>,<sup>94</sup> 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-C<sub>3</sub>N<sub>4</sub> (Fig. 1A). The e in the CB of semiconductor 1 have a tendency to migrate to the CB of g-C<sub>3</sub>N<sub>4</sub> owing to the band alignment. Similarly, the h<sup>+</sup> in the VB of g-C<sub>3</sub>N<sub>4</sub> easily migrate to the VB of semiconductor 1.96,97 Owing to the e and h being concentrated on g-C<sub>3</sub>N<sub>4</sub> and semiconductor 1, respectively, high separation efficiency of the photogenerated e<sup>-</sup> and h<sup>+</sup> 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<sup>+</sup> in g-C<sub>3</sub>N<sub>4</sub> to migrate to the CB of g-C<sub>3</sub>N<sub>4</sub> because of strong electrostatic repulsions, such as e<sup>-</sup>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-C<sub>3</sub>N<sub>4</sub> hybrids.<sup>73</sup> 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-C<sub>3</sub>N<sub>4</sub> 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<sup>+</sup> in g-C<sub>3</sub>N<sub>4</sub> 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-C<sub>3</sub>N<sub>4</sub> to the VB of semiconductor 1 with adequate photogenerated h<sup>+</sup> is very favorable owing to the effective electrostatic attraction.72 It is believed that g-C3N4-based Z-scheme systems exhibit faster charge transfer<sup>101</sup> when compared with g-C<sub>3</sub>N<sub>4</sub>-based type-II heterojunctions.

Until now, there have been two main types of g-C<sub>3</sub>N<sub>4</sub>based Z-scheme systems, including g-C<sub>3</sub>N<sub>4</sub>-based direct Z-scheme (Fig. 1B) without an electron mediator and g-C<sub>3</sub>N<sub>4</sub>-based semiconductor-conductor-semiconductor (SCS)

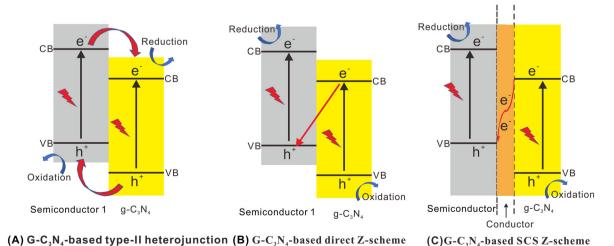


Fig. 1 Comparative analysis of the charge transfer mechanism on g-C<sub>3</sub>N<sub>4</sub>-based type-II heterojunctions (A), g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme (B) and (C) g-C<sub>3</sub>N<sub>4</sub>-based SCS Z-scheme systems.

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Z-scheme system (Fig. 1C) with an electron mediator (conductor), e.g., conductive polymer, graphene, noble metal. The g-C<sub>3</sub>N<sub>4</sub>-based direct Z-scheme systems (e.g., TiO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub>,<sup>73</sup> ZnO/g-C<sub>3</sub>N<sub>4</sub>,<sup>108</sup> WO<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub>,<sup>109</sup> Ag<sub>3</sub>PO<sub>4</sub>/ g-C<sub>3</sub>N<sub>4</sub>,<sup>110</sup> BiVO<sub>4</sub>/g-C<sub>3</sub>N<sub>4</sub>,<sup>111</sup> Bi<sub>2</sub>WO<sub>6</sub>/g-C<sub>3</sub>N<sub>4</sub>, <sup>112</sup> BiOCl/g-C<sub>3</sub>N<sub>4</sub>, <sup>113</sup> AgBr/g-C<sub>3</sub>N<sub>4</sub> (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., WO<sub>3</sub>/Cu/gC<sub>3</sub>N<sub>4</sub>, <sup>116</sup> Ag<sub>3</sub>PO<sub>4</sub>/Ag/g-C<sub>3</sub>N<sub>4</sub>, <sup>117</sup> CdS/Au/g-C<sub>3</sub>N<sub>4</sub> (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., C<sub>3</sub>N<sub>4</sub>-Pdg-C<sub>3</sub>N<sub>4</sub>/CaFe<sub>2</sub>O<sub>4</sub>, 121 Cu<sub>2</sub>O, 119  $LaFeO_3/g-C_3N_4$ , <sup>120</sup> phosphorus nanosheets/FeSe<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> (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-C<sub>3</sub>N<sub>4</sub>-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-C<sub>3</sub>N<sub>4</sub>-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.

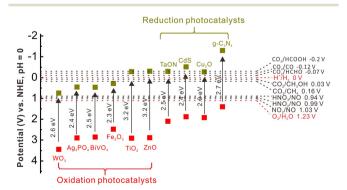


Fig. 2 Standard redox potentials of some commonly semiconductors. Reproduced with permission from ref. 35.

# 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 by investigating the photocatalytic reactions, products, and radical species.

Considering the g-C<sub>3</sub>N<sub>4</sub>/WO<sub>3</sub> photocatalysts as an example, a tight contact between g-C<sub>3</sub>N<sub>4</sub> and WO<sub>3</sub> can be found (Fig. 3A), and the individual g-C<sub>3</sub>N<sub>4</sub> and WO<sub>3</sub> components exhibit low photocatalytic performance (Fig. 3B and C). Interestingly, the g-C<sub>3</sub>N<sub>4</sub>/WO<sub>3</sub> 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<sup>-</sup>-h<sup>+</sup>. The g-C<sub>3</sub>N<sub>4</sub>/WO<sub>3</sub> composites also exhibited good stability (Fig. 3E).127 If it follows type II mechanism, the photogenerated e will transfer from the CB of g-C<sub>3</sub>N<sub>4</sub> to the CB of WO<sub>3</sub>. In this way, the g-C<sub>3</sub>N<sub>4</sub>/WO<sub>3</sub> 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-C<sub>3</sub>N<sub>4</sub>/CoTiO<sub>3</sub> direct heterojunctions through analyzing photocatalytic products. The single CoTiO<sub>3</sub> displayed very low photocatalytic activity due to its low reduction ability, but the single g-C<sub>3</sub>N<sub>4</sub> 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-C<sub>3</sub>N<sub>4</sub>/CoTiO<sub>3</sub> 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-C<sub>3</sub>N<sub>4</sub>/CoTiO<sub>3</sub> exhibited enhanced photocatalytic property, suggesting that the g-C<sub>3</sub>N<sub>4</sub>/CoTiO<sub>3</sub> 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

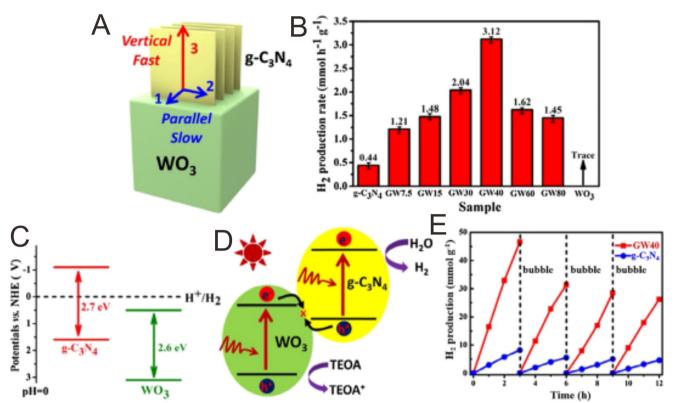


Fig. 3 (A) The growth and extension of  $g-C_3N_4$  parallel with and perpendicular to the flat surface of the WO<sub>3</sub> nanocuboids. (B) Comparison of the photocatalytic  $H_2$  production properties. (C) Band structure of  $g-C_3N_4$  and  $WO_3$ . (D) The direct Z-scheme charge migration path in the  $g-C_3N_4$ WO<sub>3</sub> composites. (E) The photocatalytic stability tests of the g-C<sub>3</sub>N<sub>4</sub> and g-C<sub>3</sub>N<sub>4</sub>/WO<sub>3</sub> 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-workers<sup>130</sup> proposed Fe<sub>2</sub>O<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub> photocatalysts, which showed improved photocatalytic performance when compared with the pure Fe<sub>2</sub>O<sub>3</sub> and g-C<sub>3</sub>N<sub>4</sub> components. The TEM image (Fig. 4) showed that Pt nanoparticles were deposited on g-C<sub>3</sub>N<sub>4</sub>, but not on Fe<sub>2</sub>O<sub>3</sub>, suggesting that the photogenerated e mainly focused on g-C<sub>3</sub>N<sub>4</sub>. If there were no close contacts between g-C<sub>3</sub>N<sub>4</sub> and Fe<sub>2</sub>O<sub>3</sub>, Pt nanoparticles would randomly photodeposit on g-C<sub>3</sub>N<sub>4</sub> and

Fe<sub>2</sub>O<sub>3</sub>. If it followed the type II mechanism, the Pt nanoparticles would mainly concentrate on Fe<sub>2</sub>O<sub>3</sub>. Instead, the Pt nanoparticles mainly concentrated on g-C<sub>3</sub>N<sub>4</sub>. Therefore, the Fe<sub>2</sub>O<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub> systems should follow the Z-scheme mechanism.

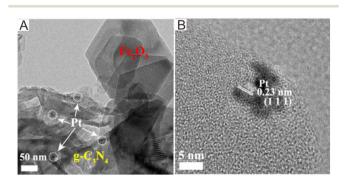


Fig. 4 (A) TEM image and (B) HRTEM image of the Fe<sub>2</sub>O<sub>3</sub>/q-C<sub>3</sub>N<sub>4</sub> composites after photodeposition of Pt nanoparticles. Reproduced with permission from ref. 130.

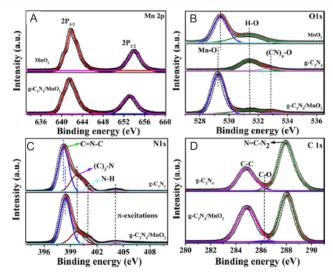


Fig. 5 XPS spectra of (A) Mn 2p, (B) O 1s, (C) N 1s, and (D) C 1s for g-C<sub>3</sub>N<sub>4</sub>, MnO<sub>2</sub>, and g-C<sub>3</sub>N<sub>4</sub>/MnO<sub>2</sub>. Reproduced with permission from ref.

#### 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-C<sub>3</sub>N<sub>4</sub>/MnO<sub>2</sub>, 137 the Mn 2p spectrum of the MnO<sub>2</sub> 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-C<sub>3</sub>N<sub>4</sub> sample only exhibited the adsorbed oxygen species and other oxygen impurities (Fig. 5B). In view of g-C<sub>3</sub>N<sub>4</sub>/MnO<sub>2</sub>, 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-C<sub>3</sub>N<sub>4</sub> (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<sup>2</sup>-hybridized nitrogen (C=N-C), bridging nitrogen ((C)<sub>3</sub>-N), and N-H bonding in the g-C<sub>3</sub>N<sub>4</sub> framework, respectively. When compared with the MnO<sub>2</sub> 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-C<sub>3</sub>N<sub>4</sub> to MnO<sub>2</sub> 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-workers<sup>144</sup> reported a g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme system through encapsulation of g-C<sub>3</sub>N<sub>4</sub> into hollow In<sub>2</sub>O<sub>3</sub> 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-C<sub>3</sub>N<sub>4</sub> 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.

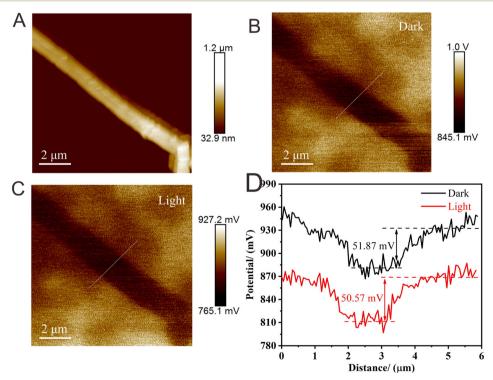


Fig. 6 (A) Atomic force microscopy image of the samples. (B) KPFM potential image of the samples under dark conditions. (C) KPFM potential image of the samples under light illumination. (D) The corresponding surface curves correspond to the white light. Reproduced with permission from ref. 144.

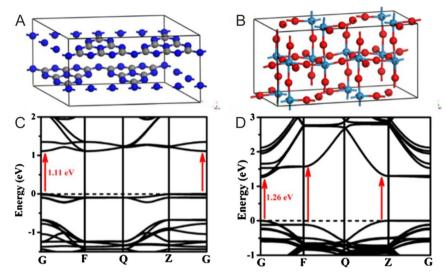


Fig. 7 Crystal structures of (A) g-C<sub>3</sub>N<sub>4</sub> and (B) monoclinic WO<sub>3</sub>. The calculated electronic band structures of (C) g-C<sub>3</sub>N<sub>4</sub> and (D) WO<sub>3</sub>. Reproduced with permission from ref. 127.

#### 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<sup>+</sup> to the e<sup>-</sup> 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,  $m^*$ ,  $\nu$ , 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-C<sub>3</sub>N<sub>4</sub> and WO<sub>3</sub>

Species	Effective mass	<i>G–F</i> direction	<i>G–Z</i> direction	<i>F–Q</i> direction	<i>Z</i> – <i>Q</i> direction
g-C <sub>3</sub> N <sub>4</sub>	m*	0.72	71	_	_
	$m_{\mathrm{e}}^{*}$	0.53	1.8	_	_
	D	1.36	39.44		_
$WO_3$	$m_{ m h}^*$	0.53	_	0.49	0.51
	$m_{\mathrm{e}}^{*}$	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<sup>+</sup> to the e<sup>-</sup>, and the effective mass of photogenerated h<sup>+</sup> and e<sup>-</sup>.

The calculated effective masses of photogenerated e and h<sup>+</sup> 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-C<sub>3</sub>N<sub>4</sub>, the excited electrons of WO<sub>3</sub> exhibit a higher transfer efficiency from bulk to interface, and then transfer into g-C<sub>3</sub>N<sub>4</sub>. In this way, the interfacial electron transfer path in g-C<sub>3</sub>N<sub>4</sub>/WO<sub>3</sub> occurs from WO<sub>3</sub> to g-C<sub>3</sub>N<sub>4</sub>, 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-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts, and selecting a suitable method is crucial. In the following sections, different preparation methods for g-C<sub>3</sub>N<sub>4</sub>-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-C<sub>3</sub>N<sub>4</sub>-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-C<sub>3</sub>N<sub>4</sub> suspension, and then vigorously stirred to ensure full adsorption of Ag+ on the g-C<sub>3</sub>N<sub>4</sub> surface. Subsequently, the Cl<sup>-</sup> ions and Ag<sup>+</sup> ions were fully coated on the g-C<sub>3</sub>N<sub>4</sub> 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	<ul><li>(i) Facile, eco-friendly, and highly efficient</li><li>(ii) Produced photocatalysts with a relatively high crystallinity, narrow size distribution, and controllable morphology</li></ul>	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-C<sub>3</sub>N<sub>4</sub> direct Z-scheme photocatalysts. The coating of AgCl on g-C<sub>3</sub>N<sub>4</sub> was proved via XRD and FTIR spectra, which also confirmed the close interface between AgCl and g-C<sub>3</sub>N<sub>4</sub> 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-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts without further post-treatment. Liu and co-workers<sup>156</sup> reported on Bi<sub>2</sub>WO<sub>6</sub> quantum dots/g-C<sub>3</sub>N<sub>4</sub> binary Z-scheme systems via a simple one-pot hydrothermal strategy. In this work, porous g-C<sub>3</sub>N<sub>4</sub> ultrathin nanosheets provided a high specific surface area for decorating Bi<sub>2</sub>WO<sub>6</sub> quantum dots, leading to a strong interaction between the Bi<sub>2</sub>WO<sub>6</sub> quantum dots and g-C<sub>3</sub>N<sub>4</sub> 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 milling<sup>159</sup> 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-workers<sup>161</sup> explored the effect of calcination temperature on the preparation of hierarchical TiO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> Z-scheme photocatalysts. The insertion of g-C<sub>3</sub>N<sub>4</sub>

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-workers<sup>165</sup> prepared Cd<sub>x</sub>Zn<sub>1-x</sub>S/Au/g-C<sub>3</sub>N<sub>4</sub> 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 TiO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub>/RGO ternary Z-scheme heterojunction via an electrospinning coupled approach. The crystal structure, morphology, composition, specific surface

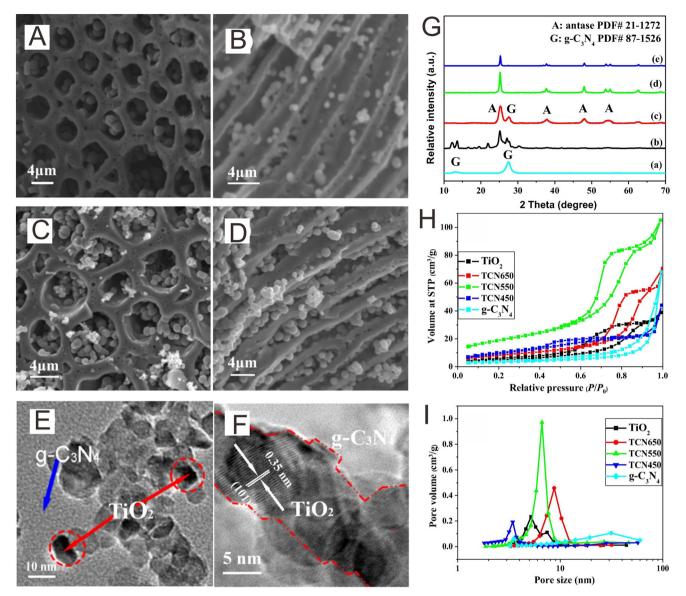


Fig. 8 SEM images at different perspective views of (A and B) the pure hierarchical TiO<sub>2</sub> framework and (C and D) the TiO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> Z-scheme photocatalyst calcination at 550 °C (denoted as TCN550). TEM (E) and HRTEM (F) images of the TCN550. XRD (G), N2 adsorption 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-workers<sup>175</sup> proposed a simple electrostatic selfassembly approach for preparing g-C<sub>3</sub>N<sub>4</sub>/ZnO Z-scheme photocatalysts. In this system, the electrostatic force between g-C<sub>3</sub>N 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-C<sub>3</sub>N<sub>4</sub> suspension, the ZnO nanoplates' surface was spontaneously coated with g-C<sub>3</sub>N<sub>4</sub> nanosheets to minimize the surface energy, resulting in the formation of a core-shell N-doped ZnO/g-C<sub>3</sub>N<sub>4</sub> 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

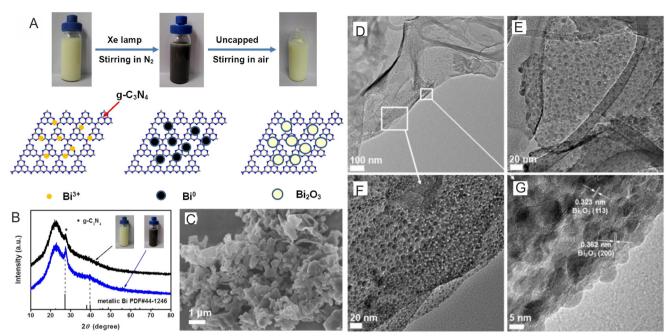


Fig. 9 (A) Diagrammatic sketch of the preparation of Bi<sub>2</sub>O<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub>. (B) XRD patterns of the intermediate products. (C) FESEM image of Bi<sub>2</sub>O<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub>. C<sub>3</sub>N<sub>4</sub>. TEM (D-F) and HRTEM (G) images of Bi<sub>2</sub>O<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub>. Reproduced with permission from ref. 180.

promising for preparing Z-scheme photocatalysts. A Bi<sub>2</sub>O<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub> 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 Bi<sup>3+</sup> to Bi<sub>2</sub>O<sub>3</sub> (Fig. 9A). The uniform dispersion of quantum-sized Bi<sub>2</sub>O<sub>3</sub> on the g-C<sub>3</sub>N<sub>4</sub> 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 BiVO<sub>4</sub>(040)-Ag@CdS Z-scheme photocatalyst in which Ag was selectively photo-deposited on the (040) facets of BiVO<sub>4</sub>, and the CdS nanorods were decorated on BiVO<sub>4</sub>(040)-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.<sup>186</sup> Ohno and coworkers<sup>187</sup> fabricated three kinds of g-C<sub>3</sub>N<sub>4</sub>/WO<sub>3</sub> Z-scheme photocatalysts via grinding, impregnation assisted with sonication, and planetary ballmilling methods. Different from the agglomerated WO3 decorated on the g-C<sub>3</sub>N<sub>4</sub> nanoplate prepared through the first two methods, WO<sub>3</sub> 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

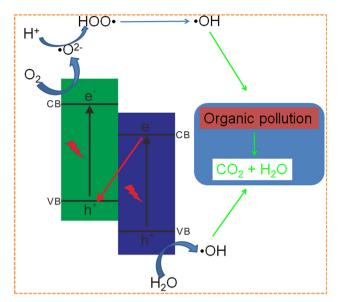


Fig. 10 Diagrammatic explanation of organic pollutant degradation over q-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts.

have recently been excessively discharged to soil and water, which have negative influences on the environment and human health. 193-201 g-C<sub>3</sub>N<sub>4</sub>-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-C<sub>3</sub>N<sub>4</sub>/TiO<sub>2</sub> 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 photocatalytic properties by  $g-C_3N_4/TiO_2$ Z-scheme photocatalysts. Dong and co-workers<sup>205</sup> fabricated an allsolid-state (ASS) Z-scheme photocatalyst through decorating g-C<sub>3</sub>N<sub>4</sub> nanosheets on TiO<sub>2</sub> 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 coworkers<sup>206</sup> prepared the g-C<sub>3</sub>N<sub>4</sub> quantum dots (QDs)-modified rutile TiO<sub>2</sub> (rTiO<sub>2</sub>) (g-C<sub>3</sub>N<sub>4</sub> QDs-rTiO<sub>2</sub>) 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-workers<sup>208</sup> fabricated a g-C<sub>3</sub>N<sub>4</sub>/TiO<sub>2</sub>/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 group<sup>209</sup> studied the influence of the exposed {001} and {101} facets of TiO2 on the photocatalytic activity. They first fabricated TiO<sub>2</sub> nanosheets (NSs) and TiO<sub>2</sub> nanoboxes (NBs), and then combined them with g-C<sub>3</sub>N<sub>4</sub> to prepare TiO<sub>2</sub> NS/g-C<sub>3</sub>N<sub>4</sub> and TiO<sub>2</sub> NBs/g-C<sub>3</sub>N<sub>4</sub>, 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 TiO<sub>2</sub> NBs came into contact with g-C<sub>3</sub>N<sub>4</sub> (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<sup>+</sup> 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-C<sub>3</sub>N<sub>4</sub>/WO<sub>3</sub> 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-C<sub>3</sub>N<sub>4</sub> to construct Z-scheme photocatalysts. The Chen group<sup>213</sup> developed a WO<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub> Z-scheme photocatalyst. The reaction rate constants for degrading MB and BF over WO<sub>3</sub>(5.0 wt%)/g-C<sub>3</sub>N<sub>4</sub> were found to be 4.77 and 3.84 times greater than that of the bare g-C<sub>3</sub>N<sub>4</sub>, respectively. The Jia group<sup>214</sup> developed a new bifunctional WO<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub> Z-scheme photocatalyst by in situ liquid phase strategy. The photocatalytic degradation efficiency of the 30 wt%WO<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub> was found to be 2.5 and 2.7 times higher than the individual g-C<sub>3</sub>N<sub>4</sub> and WO<sub>3</sub>, respectively. Kang and co-workers<sup>215</sup> fabricated WO<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub> 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-workers<sup>216</sup> constructed a WO<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub> Z-scheme photocatalyst through in situ acidic precipitation and subsequent calcination processes, which also showed improved photocatalytic property for RhB degradation. Zheng and co-workers<sup>217</sup> reported a C-doped g-C<sub>3</sub>N<sub>4</sub>/WO<sub>3</sub> 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}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-g_{5}$-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 <sub>3</sub> N <sub>4</sub> /TiO <sub>2</sub>	Calcination route	300	15 W UV lamp, 365 nm	HCHO/170 ppm	60 (~94)	7.36	73
g-C <sub>3</sub> N <sub>4</sub> /TiO <sub>2</sub>	Electrochemical route	_	300 W xenon lamp, >420 nm	RhB/5 mg L <sup>-1</sup>	300 (~77)	4.48	205
g-C <sub>3</sub> N <sub>4</sub> /rTiO <sub>2</sub>	Calcination route	50	500 W xenon lamp, >420 nm	RhB/5 mg L <sup>-1</sup>	240 (~98)	11.5	206
g-C <sub>3</sub> N <sub>4</sub> /TiO <sub>2</sub> /CuO	Hydrothermal strategy	50	500 W xenon lamp	$RhB/0.5 \text{ mg L}^{-1}$	120 (~97)	0.95	208
g-C <sub>3</sub> N <sub>4</sub> /TiO <sub>2</sub> -HNB	Microwave-assisted	_	3 W LED lamp,	Brilliant red X-3B/1.0	45 (~99)	5.1	209
	strategy		365 nm	$\times$ 10 <sup>-4</sup> mmol L <sup>-1</sup>			
$WO_3/g$ - $C_3N_4$	Ball milling and heat	10	500 W xenon	MB and BF/0.9 $\times$ 10 <sup>-5</sup>	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 mg L <sup>-1</sup>	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 <sup>-1</sup>	120 (~90)	_	215
C-doped g-C <sub>3</sub> N <sub>4</sub> /WO <sub>3</sub>	Cellulose-assisted	100	500 W xenon	$TC/10 \text{ mg L}^{-1}$	60 (~70)	_	217
WO <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub> /Bi <sub>2</sub> O <sub>3</sub>	strategy One step co-calcination	100	lamp, >420 nm 300 W xenon	$TC/10 \text{ mg L}^{-1}$	60 (~80)	2.37	159
WO3/g-C3IV4/DI2O3	way	100	lamp, >420 nm	TO/TO HIG L	00 (*-80)	2.37	133
g-C <sub>3</sub> N <sub>4</sub> /MnO <sub>2</sub>	Wet-chemical method	50	300 W xenon lamp	$RhB/10 \text{ mg L}^{-1}$	180 (~77)	3.3	137
$\text{Bi}_2\text{O}_3/\text{g-C}_3\text{N}_4$	Ball milling and heat	5	500 W xenon	MB and RhB/1.1 × $10^{-5}$	MB 120 (~76)	2.53 and	223
2 3 0 3 4	treatment methods		lamp, 470 nm	and $1.0 \times 10^{-5} \text{ mol L}^{-1}$	RhB 120 (~73)	1.01	
g-C <sub>3</sub> N <sub>4</sub> /Bi <sub>2</sub> O <sub>3</sub> /BiPO <sub>4</sub>	Calcination route	15		$MO/10 \text{ mg L}^{-1}$	160 (~88)	1.15	225
β-Bi <sub>2</sub> O <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>	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 <sub>3</sub> N <sub>4</sub> /Bi <sub>2</sub> WO <sub>6</sub>	Mixing and heating method	50	300 W xenon lamp, 350–780 nm	2,4-DCP/20 mg $L^{-1}$	120 (>99)	1.13	228
$g$ - $C_3N_4/Bi_2WO_6$	One-pot hydrothermal	50	Xenon lamp	RhB/10 mg $L^{-1}$	30 (>99)	8.17	156
g-C <sub>3</sub> N <sub>4</sub> /RGO/Bi <sub>2</sub> WO <sub>6</sub>	strategy Hydrothermal method	250	(XL-300, Yirda) 500 W xenon	TCP/20–100 mg $L^{-1}$	120 (>99)	3.15	229
Ag-decorated	Polymerization method	40	lamp, >420 nm 300 W xenon	RhB/10 mg L <sup>-1</sup>	90 (>99)	_	230
$g$ - $C_3N_4/Bi_2WO_6$ $Bi_3O_4Cl/g$ - $C_3N_4$	Solid-phase calcination	50	lamp, 400 nm 250 W xenon	RhB and TC/10 and 10	RhB 60 (>98) TC	— and 2.05	231
	method		lamp, >420 nm	mg L <sup>-1</sup>	60 (>76)		
BiOBr/RGO/pg-C <sub>3</sub> N <sub>4</sub>	Solvothermal method	50	300 W xenon	RhB and TC/10 and 10	RhB 7.5 (>99) TC	— and —	232
Bi <sub>12</sub> GeO <sub>20</sub> /g-C <sub>3</sub> N <sub>4</sub>	Sol-gel method	300	lamp, 420 nm 300 W Dy lamp,	mg L <sup>-1</sup> RhB/5 mg L <sup>-1</sup>	60 (>97) 50 (>99)	2.55	233
g-C <sub>3</sub> N <sub>4</sub> /C@Bi <sub>2</sub> MoO <sub>6</sub>	Hydrothermal deposition	100	400 nm 500 W xenon	β-Naphthol/15 mg L <sup>-1</sup>	150 (>62)	0.63	234
	method		lamp, >420 nm				
$Ag_3PO_4/g$ - $C_3N_4$	<i>In situ</i> precipitation method	5	300 W xenon lamp, >400 nm	SMX/1 mg L <sup>-1</sup>	10 (~98)	6.3	238
$Ag_3PO_4/g-C_3N_4$	<i>In situ</i> precipitation method	30	300 W xenon lamp	$MO/10 \text{ mg L}^{-1}$	5 (>99)	_	239
g-C <sub>3</sub> N <sub>4</sub> /Ag <sub>2</sub> WO	In situ precipitation	30	300 W xenon lamp	$MO/10~mg~L^{-1}$	150 (~98)	_	240
$Ag_2CrO_4/g$ - $C_3N_4$	method Precipitation method	20	300 W xenon	MO and RhB/10 and 10	MO 21 (~90) RhB	9.64 and	241
Ag@AgBr/g-C <sub>3</sub> N <sub>4</sub>	Deposition-precipitation	100	lamp, >400 nm 300 W xenon lamp	mg L <sup>-1</sup> MO and MB/10 and 10	40 (~97) MO 10 (~97) RhB	9.21 — and —	117
Ag <sub>2</sub> CrO <sub>4</sub> /Ag/g-C <sub>3</sub> N <sub>4</sub>	method <i>In situ</i> growth and	50	500 W xenon	mg L <sup>-1</sup> 2,4-DCP/10 mg L <sup>-1</sup>	10 (~96) 120 (~96)	1.52	243
	photoreduction methods		lamp, <420 nm	-1-1-1-1			
g-C <sub>3</sub> N <sub>4</sub> /Ag/MoS <sub>2</sub>	In situ growth and precipitation methods	100	300 W xenon lamp, >420 nm	RhB/20 mg L <sup>-1</sup>	60 (>99)	_	247
g-C <sub>3</sub> N <sub>4</sub> /ZnO	Ultrasonically exfoliated method	100	300 W xenon lamp, >420 nm	$4-CP/10^{-4} \text{ mol L}^{-1}$	90 (>99)	5.09	248
CdS/RGO/g-C <sub>3</sub> N <sub>4</sub>	Hydrothermal method	20	350 W xenon lamp	Atrazine/—	300 (~92)	0.71	249
g-C <sub>3</sub> N <sub>4</sub> -RGO-TiO <sub>2</sub>	Liquid-precipitation	50	300 W xenon	RhB/30 mg L <sup>-1</sup>	180 (~92)	1.37	250
0 0 4 2	strategy		lamp, >420 nm	, - · · · · · · · · · · · · · · · · · ·	()		
	Calcination method	30	350 W xenon	$TC/20 \text{ mg L}^{-1}$	90 (~90)	2.23	251
$MoO_3/g-C_3N_4$	Calcination method		lamp, >420 nm		,		

Table 3 (continued)

Photocatalysts	Preparation strategy	Mass (mg)	Light source	Target pollutant/initial concentration	Degradation time (min) and rate (%)	$K_{\text{app}}^{a}$ $[10^{-2} \text{ min}^{-1}]$	Ref.
NiTiO <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>	Calcination method	40	30 W visible LED	RhB/10 mg L <sup>-1</sup>	_	3.09	253
LaFeO <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>	Calcination method	30	300 W xenon lamp, >420 nm	RhB/25 mg L <sup>-1</sup>	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 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 <sup>-1</sup>	100 (~96)	0.57	255
$CDs/g$ - $C_3N_4/SnO_2$	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 <sup>-1</sup>	240 (>99)	1.39	246

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 WO<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub> composites were mainly because of the unusual merits of the Z-scheme photocatalytic systems.

### 5.3. g-C<sub>3</sub>N<sub>4</sub>/MnO<sub>2</sub> Z-scheme photocatalysts

<sup>a</sup> K<sub>app</sub> represents the reaction rate constant.

MnO2 is gradually becoming recognized as an efficient photocatalyst owing to its excellent photocatalytic performance, suitable bandgap value, and optical stability.<sup>218</sup> Xu and co-workers<sup>137</sup> prepared g-C<sub>3</sub>N<sub>4</sub>/MnO<sub>2</sub> Z-scheme photocatalysts. There were many functional groups and exposed lone pair electrons from N atoms on the g-C<sub>3</sub>N<sub>4</sub> surfaces, which provided abundant adsorption sites for Mn<sup>2+</sup> ions (Fig. 12A). It should be noted that the thicknesses of the g-C<sub>3</sub>N<sub>4</sub> nanosheets, MnO<sub>2</sub> nanosheets, and the g-C<sub>3</sub>N<sub>4</sub>/MnO<sub>2</sub> hybrid were approximately 3.4, 2.2, and 5.7 nm, respectively, which were roughly equal to the stacking distance of the g-C<sub>3</sub>N<sub>4</sub> and MnO<sub>2</sub> nanosheets (Fig. 12B-D). The electrostatic potentials of MnO<sub>2</sub> (001) and g-C<sub>3</sub>N<sub>4</sub> (001) were measured to be 6.8 and 4.5 eV, respectively, suggesting that the electrons on the g-C<sub>3</sub>N<sub>4</sub> will spill onto MnO<sub>2</sub> via the interface (Fig. 12E-G). Thus, g-C<sub>3</sub>N<sub>4</sub> was more positively charged. Photocatalytic experiments suggested that the prepared g-C<sub>3</sub>N<sub>4</sub>/MnO<sub>2</sub> exhibited better photocatalytic property when compared with the bare  $g\text{-}C_3N_4$  and  $MnO_2$  (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-C<sub>3</sub>N<sub>4</sub>/ MnO<sub>2</sub> should obey the direct Z-scheme mechanism (Fig. 12J and K). Moreover, the positive slopes of the Mott-Schottky plots indicated that both g-C<sub>3</sub>N<sub>4</sub> and MnO<sub>2</sub> 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-C<sub>3</sub>N<sub>4</sub> and MnO<sub>2</sub> 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-C<sub>3</sub>N<sub>4</sub>/MnO<sub>2</sub> hybrids. As for the individual g-C<sub>3</sub>N<sub>4</sub> or MnO<sub>2</sub>, the photogenerated h<sup>+</sup> in g-C<sub>3</sub>N<sub>4</sub> cannot oxidize OH<sup>-</sup> 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 MnO<sub>2</sub> will migrate to the VB of g-C<sub>3</sub>N<sub>4</sub> and then combine with the h<sup>+</sup>. The Z-scheme configuration can explain the enhanced photocatalytic property well (Fig. 13O).

### 5.4. g-C<sub>3</sub>N<sub>4</sub>/bismuth oxide Z-scheme photocatalysts

Recently, multifarious bismuth-based semiconductors, e.g.,  $Bi_2O_3$ ,  $^{159,180,223-225}$   $\beta$ - $Bi_2O_3$ ,  $^{226}$   $Bi_4O_7$ ,  $^{227}$   $Bi_2WO_6$ ,  $^{156,228-230}$   $Bi_3$ - $O_4Cl$ ,  $^{231}$  BiOBr,  $^{232}$  Bi $_{12}$ GeO $_2$ ,  $^{233}$  Bi $_2$ MoO $_6$ ,  $^{234}$  have been reported for combining with g-C<sub>3</sub>N<sub>4</sub> 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 Bi<sub>2</sub>O<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub> photocatalyst. The reaction rate constants of Bi<sub>2</sub>O<sub>3</sub> (1.0 wt%)/g-C<sub>3</sub>N<sub>4</sub> for MB and RhB degradation were approximately 3.4 and 5 times greater that of the bare g-C<sub>3</sub>N<sub>4</sub> and Bi<sub>2</sub>O<sub>3</sub>, 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-workers<sup>227</sup> fabricated a

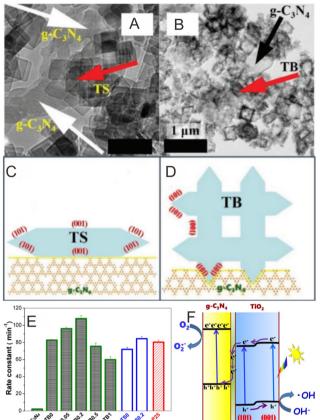


Fig. 11 TEM images of (A) TiO<sub>2</sub> NSs/g-C<sub>3</sub>N<sub>4</sub> and (B) TiO<sub>2</sub> NBs/g-C<sub>3</sub>N<sub>4</sub>. Diagrammatic sketch of the contact interface between TiO2 and g- $C_3N_4$  on (C)  $TiO_2$   $NSs/g-C_3N_4$  and (D)  $TiO_2$   $NBs/g-C_3N_4$ . (E) Comparison of the photocatalytic 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 TiO<sub>2</sub>/g-C<sub>3</sub>N<sub>4</sub> hybrids. Reproduced with permission from ref. 209.

high-performance g-C<sub>3</sub>N<sub>4</sub>/Bi<sub>4</sub>O<sub>7</sub> Z-scheme photocatalyst via a simple way. In contrast to the single photocatalysts, the asprepared g-C<sub>3</sub>N<sub>4</sub>/Bi<sub>4</sub>O<sub>7</sub> 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-workers<sup>228</sup> reported a simple mixing and heating strategy to fabricate the g-C<sub>3</sub>N<sub>4</sub>/Bi<sub>2</sub>WO<sub>6</sub> Z-scheme photocatalyst. The optimal dechlorination rate constant of 2,4-DCP over g-C<sub>3</sub>N<sub>4</sub>/Bi<sub>2</sub>WO<sub>6</sub> was 1.13 h<sup>-1</sup>, exceeding those of the bare g-C<sub>3</sub>N<sub>4</sub> and Bi2WO<sub>6</sub> by 7.6 and 8.9 times, respectively. Yin and co-workers<sup>233</sup> prepared a new ASS  $Bi_{12}GeO_{20}/g-C_3N_4$ Z-scheme photocatalyst. The obtained Bi<sub>12</sub>GeO<sub>20</sub>/g-C<sub>3</sub>N<sub>4</sub> photocatalyst displayed improved photocatalytic property for degrading microcystin-LR and RhB in contrast to the individual Bi12-GeO<sub>20</sub> and g-C<sub>3</sub>N<sub>4</sub>. 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 gC<sub>3</sub>N<sub>4</sub> to fabricate Z-scheme photocatalysts for enhancing photocatalytic properties owing to their suitable CB and CB

# 5.5. g-C<sub>3</sub>N<sub>4</sub>/Ag-containing semiconductor Z-scheme photocatalysts

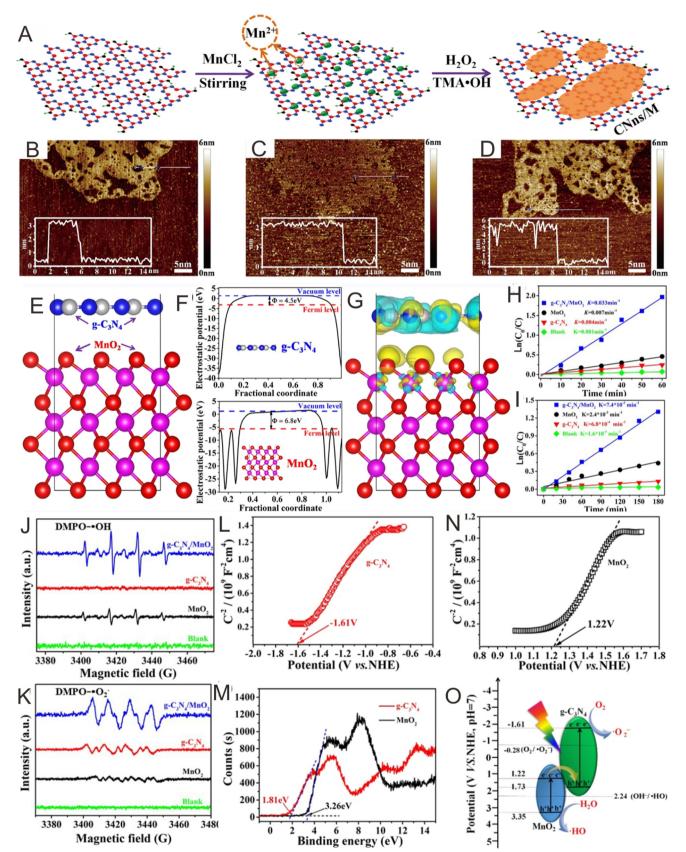
Ag-containing photocatalysts can greatly improve the photocatalytic property of g-C<sub>3</sub>N<sub>4</sub> through the Z-scheme mechanism. Some examples of Ag-containing photocatalysts with improved properties include Ag<sub>3</sub>PO<sub>4</sub>/g-C<sub>3</sub>N<sub>4</sub>, <sup>238,239</sup> Ag<sub>2</sub>-WO<sub>4</sub>/g-C<sub>3</sub>N<sub>4</sub>, <sup>240</sup> and Ag<sub>2</sub>CrO<sub>4</sub>/g-C<sub>3</sub>N<sub>4</sub>. <sup>241</sup> In particular, utilizing noble metal Ag as the electron mediator for fabrication of Agsemiconductors/g-C<sub>3</sub>N<sub>4</sub> photocatalysts can achieve more effective photocatalytic performance.<sup>242</sup> The Guo group<sup>117</sup> fabricated various Ag@AgBr/g-C<sub>3</sub>N<sub>4</sub> Z-scheme photocatalysts photoreduction of AgBr/g-C<sub>3</sub>N<sub>4</sub> hybrids. A dramatically improved photocatalytic property of Ag@AgBr/g-C<sub>3</sub>N<sub>4</sub> was achieved, and is mainly attributed to the high light utilization and Z-scheme mechanism. Quan and coworkers<sup>243</sup> prepared Ag<sub>2</sub>CrO<sub>4</sub>/Ag/g-C<sub>3</sub>N<sub>4</sub> Z-scheme photocatalysts via simple in situ growth and photoreduction strategies. Because of the strong light utilization, effective separation of the photogenerated e<sup>-</sup> and h<sup>+</sup>, and the excellent redox ability, the obtained Ag<sub>2</sub>CrO<sub>4</sub>/Ag/g-C<sub>3</sub>N<sub>4</sub> hybrids showed remarkably improved photocatalytic performance for 2,4-DCP degradation, exceeding that of the pure g-C<sub>3</sub>N<sub>4</sub> by 5.2 times.

## 5.6. Metal-free g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts

Fabricating metal-free g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts is a very promising and environmentally friendly method for degrading organic pollutants.<sup>244,245</sup> Huang and co-workers<sup>246</sup>  $constructed \quad a \quad metal\text{-}free \quad 2D \quad g\text{-}C_3N_4/g\text{-}C_3N_4 \quad Z\text{-}scheme$ 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- $C_3N_4/g$ - $C_3N_4$ Z-scheme photocatalysts showed photocatalytic performance when compared to the conventional g-C<sub>3</sub>N<sub>4</sub>/g-C<sub>3</sub>N<sub>4</sub> homojunction and bulk g-C<sub>3</sub>N<sub>4</sub>. This work provides some new design concepts for Z-scheme systems with specific 2D morphology for enhanced organic pollutant removal.

### 5.7. g-C<sub>3</sub>N<sub>4</sub>-based SCS Z-scheme photocatalysts

As mentioned above, the g-C<sub>3</sub>N<sub>4</sub>-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.<sup>247</sup> They prepared a Z-scheme g-C<sub>3</sub>N<sub>4</sub>/Ag/MoS<sub>2</sub> ternary plasmonic Z-scheme photocatalyst with flowerlike nanostructures with a diameter of  $\sim 0.4 - 0.6$ (Fig. The prepared g-C<sub>3</sub>N<sub>4</sub>/Ag/MoS<sub>2</sub>



 $\textbf{Fig. 12} \hspace{0.2cm} \textbf{(A)} \hspace{0.2cm} \textbf{Diagrammatic} \hspace{0.2cm} \textbf{sketch} \hspace{0.2cm} \textbf{of} \hspace{0.2cm} \textbf{g-C}_3\textbf{N}_4/\textbf{MnO}_2 \hspace{0.2cm} \textbf{prepared} \hspace{0.2cm} \textbf{through} \hspace{0.2cm} \textbf{\textit{in}} \hspace{0.2cm} \textbf{\textit{situ}} \hspace{0.2cm} \textbf{\textit{growth}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{MnO}}_2 \hspace{0.2cm} \textbf{\textit{on}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{g-C}}_3\textbf{N}_4 \hspace{0.2cm} \textbf{\textit{nanosheets.}} \hspace{0.2cm} \textbf{\textit{AFM}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{g-C}}_3\textbf{\textit{N}}_4 \hspace{0.2cm} \textbf{\textit{nanosheets.}} \hspace{0.2cm} \textbf{\textit{AFM}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{g-C}}_3\textbf{\textit{N}}_4 \hspace{0.2cm} \textbf{\textit{nanosheets.}} \hspace{0.2cm} \textbf{\textit{AFM}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{g-C}}_3\textbf{\textit{N}}_4 \hspace{0.2cm} \textbf{\textit{nanosheets.}} \hspace{0.2cm} \textbf{\textit{AFM}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{the}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm} \textbf{\textit{images}} \hspace{0.2cm} \textbf{\textit{of}} \hspace{0.2cm}$ 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-'O<sup>2-</sup> 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 MnO<sub>2</sub> and g-C<sub>3</sub>N<sub>4</sub> nanosheets. (O) Photocatalytic mechanism of  $g-C_3N_4/MnO_2$ . Reproduced with permission from ref. 137.

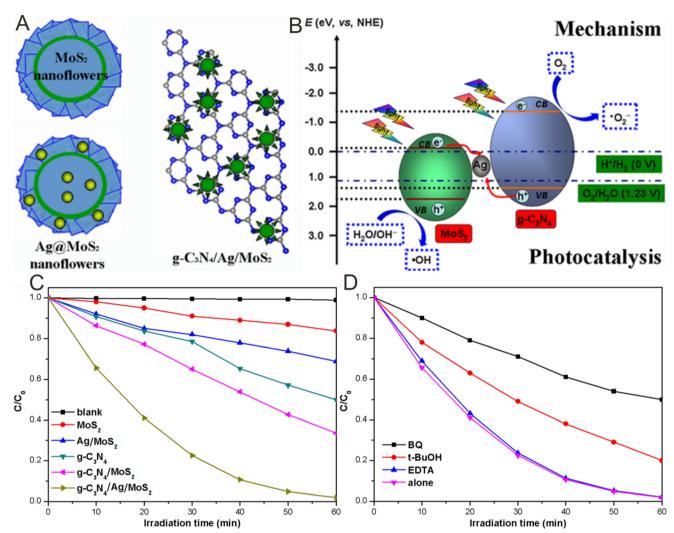


Fig. 13 (A) The nanostructures of Ag/MoS2 and g-C3N4/Ag/MoS2. (B) Diagrammatic sketch for the separation and migration of the photogenerated charges over g-C<sub>3</sub>N<sub>4</sub>/Ag/MoS<sub>2</sub>. (C and D) Photocatalytic property of the fabricated MoS<sub>2</sub>, Ag/MoS<sub>2</sub>, g-C<sub>3</sub>N<sub>4</sub>, g-C<sub>3</sub>N<sub>4</sub>/MoS<sub>2</sub>, and g-C<sub>3</sub>N<sub>4</sub>/Aq/MoS<sub>2</sub> 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-C<sub>3</sub>N<sub>4</sub>, and MoS<sub>2</sub> nanophase structures, which led Z-schememechanism-induced rapid separation of the pairs and further photogenerated  $e^--h^+$ enhanced photocatalytic property (Fig. 13B).

## 5.8. Others

In addition to the above common g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts, researchers have made many attempts to explore other candidates to fabricate g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme systems, e.g., g-C<sub>3</sub>N<sub>4</sub>/ZnO,<sup>248</sup> CdS/RGO/g-C<sub>3</sub>N<sub>4</sub>,<sup>249</sup>  $g-C_3N_4-RGO-TiO_2$ , 250  $MoO_3/g-C_3N_4$ , 251  $Cu_3P/g-C_3N_4$ , 252 NiTiO<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub>, <sup>253</sup> LaFeO<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub>, <sup>120</sup> V<sub>2</sub>O<sub>5</sub>/g-C<sub>3</sub>N<sub>4</sub>, <sup>107</sup> SnO<sub>2x</sub>/ g-C<sub>3</sub>N<sub>4</sub>, <sup>254</sup> InVO<sub>4</sub>/Bi<sub>2</sub>S<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub>, <sup>255</sup> CDs/g-C<sub>3</sub>N<sub>4</sub>/SnO<sub>2</sub>, <sup>256</sup> gC<sub>3</sub>N<sub>4</sub>/ZnFe<sub>2</sub>O<sub>4</sub>/UiO-66,<sup>256</sup> g-C<sub>3</sub>N<sub>4</sub>/BiVO<sub>4</sub>/CoFe<sub>2</sub>O<sub>4</sub>. <sup>257</sup> 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-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts<sup>258,259</sup> are also very useful in this field. The use of g-C<sub>3</sub>N<sub>4</sub>-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-C<sub>3</sub>N<sub>4</sub>-based Z-scheme systems will have enormous potential in photocatalytic organic pollutant degradation.

# 6. Conclusions and outlook

g-C<sub>3</sub>N<sub>4</sub>-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-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts can greatly boost the spatial separation of the photogenerated carrier and enhance their photocatalytic activity and light utilization. Thus, g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts exhibit great potential for organic pollution removal. Although many achievements have been obtained, the research studies on g-C<sub>3</sub>N<sub>4</sub>-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) mechanism<sup>261</sup> is more reasonable and powerful for illustrating the evidently improved photocatalytic property.
- (2) The physicochemical property of the g- $C_3N_4$ -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  $g\text{-}C_3N_4\text{-}based$  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  $g\text{-}C_3N_4\text{-}based$  Z-scheme photocatalysts is a promising method for improving their photocatalytic performance.
- (4) The photocatalytic performance of g- $C_3N_4$ -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- $C_3N_4$ -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- $C_3N_4$ -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-C<sub>3</sub>N<sub>4</sub>-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- $C_3N_4$ -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-C<sub>3</sub>N<sub>4</sub>-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-C<sub>3</sub>N<sub>4</sub>-based Z-scheme systems for organic pollution removal. Certainly, the g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme systems can achieve practical industrial applications in the near future.

# Conflicts of interest

There are no conflicts of interest to declare.

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