

Cite this: *Catal. Sci. Technol.*, 2023,  
13, 2877

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

Chunxue Li,<sup>a</sup> Hao Lu,<sup>\*b</sup> Guixiang Ding,<sup>c</sup> Qing Li<sup>\*d</sup> and Guangfu Liao <sup>\*c</sup>

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 g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts for organic pollution removal, beginning with the fundamentals of g-C<sub>3</sub>N<sub>4</sub>-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.

Received 21st February 2023,  
Accepted 24th March 2023

DOI: 10.1039/d3cy00242j

rsc.li/catalysis

### 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,

such as adsorption,<sup>7–10</sup> electrocatalysis,<sup>11,12</sup> and photocatalysis<sup>13–19</sup> (especially visible-light-responsive photocatalysis),<sup>20–24</sup> 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.<sup>19,25–31</sup> 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, suitable electronic and band structure, excellent physicochemical stability and photocatalytic property.<sup>32–47</sup> 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

<sup>a</sup> College of Ecological Environment and Urban Construction, Fujian University of Technology, Fuzhou 350118, China

<sup>b</sup> School of Materials Science and Engineering, Suzhou University of Science and Technology, Suzhou 215009, China. E-mail: haolu@usts.edu.cn

<sup>c</sup> College of Material Engineering, Fujian Agriculture and Forestry University, Fuzhou 350002, China. E-mail: liaogf@mail2.sysu.edu.cn

<sup>d</sup> Ministry of Education Key Laboratory for the Green Preparation and Application of Functional Materials, Hubei Key Laboratory of Polymer Materials, School of Materials Science and Engineering, Hubei University, Wuhan 430062, China. E-mail: liqing@hubeu.edu.cn

**Chunxue Li**

Chunxue Li obtained her B.Sc. (2016) and M.S. (2019) from Jilin Normal University. She then obtained her Ph.D. from Jiangsu University. Presently, she is a Lecturer at Fujian University of Technology. Her current research focuses on the synthesis and application of semiconductor nanomaterials for photocatalytic hydrogen evolution. She has received some awards, including the Graduate Student National Scholarship, Xue Liang

Scholarship, and Excellent Graduation Thesis of Jilin Normal University.

**Hao Lu**

Hao Lu received his Ph.D. degree in Physics from Soochow University in 2017. He is currently working at the School of Materials Science and Engineering, Suzhou University of Science and Technology, as an Associate Professor. His research interest involves the fabrication of high-performance optoelectronic devices by exploiting the morphology, surface and boundary of materials and energy bands. So far, he has published more than 60 high-

profile SCI papers, such as *Adv. Mater.*, *Adv. Funct. Mater.*, *Adv. Sci.*, *Nano Energy*, *Sci. Bull.*, and *Phys. Rev. Letters*.

production demand owing to the fast electron-hole ( $e^-$ - $h^+$ ) recombination and poor light absorption capacity.<sup>48-51</sup> Hence, it is crucial to explore new  $g$ - $C_3N_4$ -based photocatalysts with strong light absorption capacity and high  $e^-$ - $h^+$  separation for organic pollutant removal.



Guixiang Ding

*Guixiang Ding received his B.S. degree from the College of Chemical Engineering at Liaoning Normal University in 2022. His current research focuses on the study of the photocatalytic  $CO_2$  reduction and on the development of photocatalytic materials. He has published one SCI as the first author and four SCI as the second author.*

To enhance the photocatalytic property of  $g$ - $C_3N_4$ , various approaches have been proposed, *e.g.*, doping,<sup>52,53</sup> obtaining porous architectures,<sup>54,55</sup> achieving highly crystalline structures,<sup>56</sup> metal loading,<sup>57,58</sup> heterojunction construction,<sup>59-61</sup> exfoliation,<sup>62,63</sup> defect engineering,<sup>64,65</sup> and molecular and electronic structure engineering.<sup>66-69</sup> 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.<sup>70,71</sup> However, their photocatalytic activity is not significantly enhanced because of electrostatic repulsion.<sup>72</sup> To overcome this disadvantage, an emerging  $g$ - $C_3N_4$ -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.<sup>74,75</sup> 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>  $g$ - $C_3N_4$ -based Z-scheme photocatalysts have achieved increasing attention for organic pollutant removal.



Qing Li

*Qing Li obtained his B.S. (2012), M.S. (2015) and Ph.D. (2018) from Hubei University. Presently, he is an Associate Professor at Hubei University. His current research focuses on the synthesis and application of Ag-based catalytic materials. He has received some awards, including the Graduate Student National Scholarship, Excellent Innovation and Entrepreneurship Mentor, and others.*



Guangfu Liao

*Guangfu Liao received his Ph.D. degree in Material Physics & Chemistry from Sun Yat-sen University in 2020. Then, he joined the laboratory of Prof. Yi-Chun Lu at The Chinese University of Hong Kong, where he worked as a Research Associate. Subsequently, he became a Researcher at the China University of Geosciences. Now, he is a Professor at Fujian Agriculture and Forestry University. His research interests involve photo-*

*and electrocatalysis, polymer synthesis and applications, polymer membranes, redox flow batteries, nanoporous and nanostructured materials, biomaterials, gas storage and energy conversion, and others. The broader impacts of his research include polymeric photocatalysts, polymer design and synthesis, nanostructure engineering, and biomaterials. So far, he has published more than 60 high profile SCI papers, such as Matter, Progress in Materials Science, Energy & Environmental Science, Applied Catalysis B: Environmental, Nano Energy, ACS Catalysis, Chemical Science, Small, Chemical Engineering Journal, Journal of Materials Chemistry A, Journal of Catalysis, and others. The awards he has received up to now include the National Endeavor Fellowship, Guanghua Scholarship, 2022 Journal of Materials Chemistry A Emerging Investigators, and others. He also serves as an Associate Editor for Frontiers in Energy Research, Guest Editor for Polymers, and Youth Editorial Board for Advanced Fiber Materials.*

Although there have been many inspired reviews on  $g\text{-C}_3\text{N}_4$ -based photocatalysts for various applications,<sup>32,33,71,77–88</sup> a comprehensive review about  $g\text{-C}_3\text{N}_4$ -based Z-scheme photocatalysts for organic pollutant removal is nonexistent despite the great interest in this research topic. In this review, the fundamentals of  $g\text{-C}_3\text{N}_4$ -based Z-scheme photocatalysts are firstly discussed. Subsequently, the characterization strategies and preparation methods for  $g\text{-C}_3\text{N}_4$ -based Z-scheme photocatalysts are illustrated. Next, some typical  $g\text{-C}_3\text{N}_4$ -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 $g\text{-C}_3\text{N}_4$ -based Z-scheme photocatalysts?

Before introducing the  $g\text{-C}_3\text{N}_4$ -based Z-scheme photocatalysts, it is crucial to fully comprehend  $g\text{-C}_3\text{N}_4$ -based type-II heterojunctions, *e.g.*,  $\text{CeO}_2/g\text{-C}_3\text{N}_4$ ,<sup>89</sup>  $\text{Co}_3\text{O}_4/g\text{-C}_3\text{N}_4$ ,<sup>90</sup>  $\text{Cu}_2\text{O}/g\text{-C}_3\text{N}_4$ ,<sup>91</sup>  $\text{Fe}_2\text{O}_3/g\text{-C}_3\text{N}_4$ ,<sup>92</sup>  $g\text{-C}_3\text{N}_4/g\text{-C}_3\text{N}_4$ ,<sup>93</sup> h-BN/flowering  $g\text{-C}_3\text{N}_4$ ,<sup>94</sup> which display outstanding photocatalytic property owing to their high separation efficiency of photogenerated charges through the band alignment.<sup>74,95</sup> In these type-II heterojunctions, both CB and VB potentials of semiconductor 1 are lower than those of  $g\text{-C}_3\text{N}_4$  (Fig. 1A). The  $e^-$  in the CB of semiconductor 1 have a tendency to migrate to the CB of  $g\text{-C}_3\text{N}_4$  owing to the band alignment. Similarly, the  $h^+$  in the VB of  $g\text{-C}_3\text{N}_4$  easily migrate to the VB of semiconductor 1.<sup>96,97</sup> Owing to the  $e^-$  and  $h^+$  being concentrated on  $g\text{-C}_3\text{N}_4$  and semiconductor 1, respectively, high separation efficiency of the photogenerated  $e^-$  and  $h^+$  can be achieved through type-II heterojunctions.<sup>32,72,98,99</sup> However, there are still some potential problems that need to be solved. The photocatalytic activity of the charges are not

significantly enhanced.<sup>100,101</sup> It is also very difficult for the  $e^-$  in semiconductor 1 and  $h^+$  in  $g\text{-C}_3\text{N}_4$  to migrate to the CB of  $g\text{-C}_3\text{N}_4$  because of strong electrostatic repulsions, such as  $e^-e^-$  or  $h^+h^+$  interactions, respectively.<sup>74</sup> 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.<sup>71,102</sup> 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  $\text{TiO}_2/g\text{-C}_3\text{N}_4$  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.<sup>103,104</sup> The charge migration path of the direct Z-scheme photocatalysts resembles the letter “Z” (Fig. 1B), in which the  $e^-$  in  $g\text{-C}_3\text{N}_4$  with stronger oxidation ability will integrate with the  $h^+$  in semiconductor 1 with strong reduction ability.<sup>105</sup> Therefore, the  $e^-$  in semiconductor 1 with stronger reduction ability and  $h^+$  in  $g\text{-C}_3\text{N}_4$  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  $g\text{-C}_3\text{N}_4$  to the VB of semiconductor 1 with adequate photogenerated  $h^+$  is very favorable owing to the effective electrostatic attraction.<sup>72</sup> It is believed that  $g\text{-C}_3\text{N}_4$ -based Z-scheme systems exhibit faster charge transfer<sup>101</sup> when compared with  $g\text{-C}_3\text{N}_4$ -based type-II heterojunctions.

Until now, there have been two main types of  $g\text{-C}_3\text{N}_4$ -based Z-scheme systems, including  $g\text{-C}_3\text{N}_4$ -based direct Z-scheme (Fig. 1B) without an electron mediator and  $g\text{-C}_3\text{N}_4$ -based semiconductor-conductor-semiconductor (SCS) (Fig. 1C).



**Fig. 1** Comparative analysis of the charge transfer mechanism on  $g\text{-C}_3\text{N}_4$ -based type-II heterojunctions (A),  $g\text{-C}_3\text{N}_4$ -based Z-scheme (B) and (C)  $g\text{-C}_3\text{N}_4$ -based SCS Z-scheme systems.

Z-scheme system (Fig. 1C) with an electron mediator (conductor), *e.g.*, conductive polymer, graphene, noble metal.<sup>106,107</sup> 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.<sup>115</sup> As for g-C<sub>3</sub>N<sub>4</sub>-based SCS Z-scheme systems (*e.g.*, WO<sub>3</sub>/Cu/g-C<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 favorable migration of photogenerated carriers.<sup>118</sup> Moreover, the p-n Z-scheme systems (*e.g.*, C<sub>3</sub>N<sub>4</sub>-Pd-Cu<sub>2</sub>O,<sup>119</sup> LaFeO<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub>,<sup>120</sup> g-C<sub>3</sub>N<sub>4</sub>/CaFe<sub>2</sub>O<sub>4</sub>,<sup>121</sup> black 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.<sup>123</sup> The consumption of e<sup>-</sup> and h<sup>+</sup> for redox reactions should be unified. Otherwise, the accumulation of charges may lead to the self-oxidation/reduction of the photocatalysts or the mediators.<sup>124</sup>

### 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.*, 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.

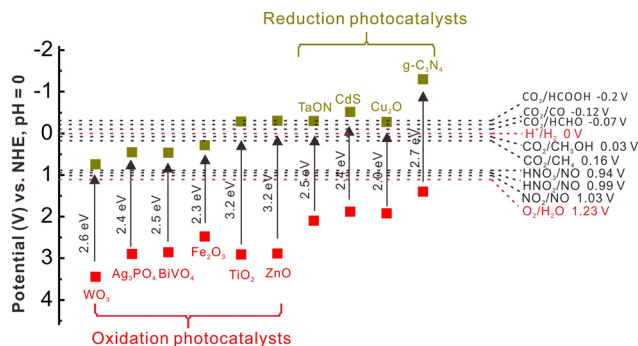


Fig. 2 Standard redox potentials of some commonly used semiconductors. Reproduced with permission from ref. 35.

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

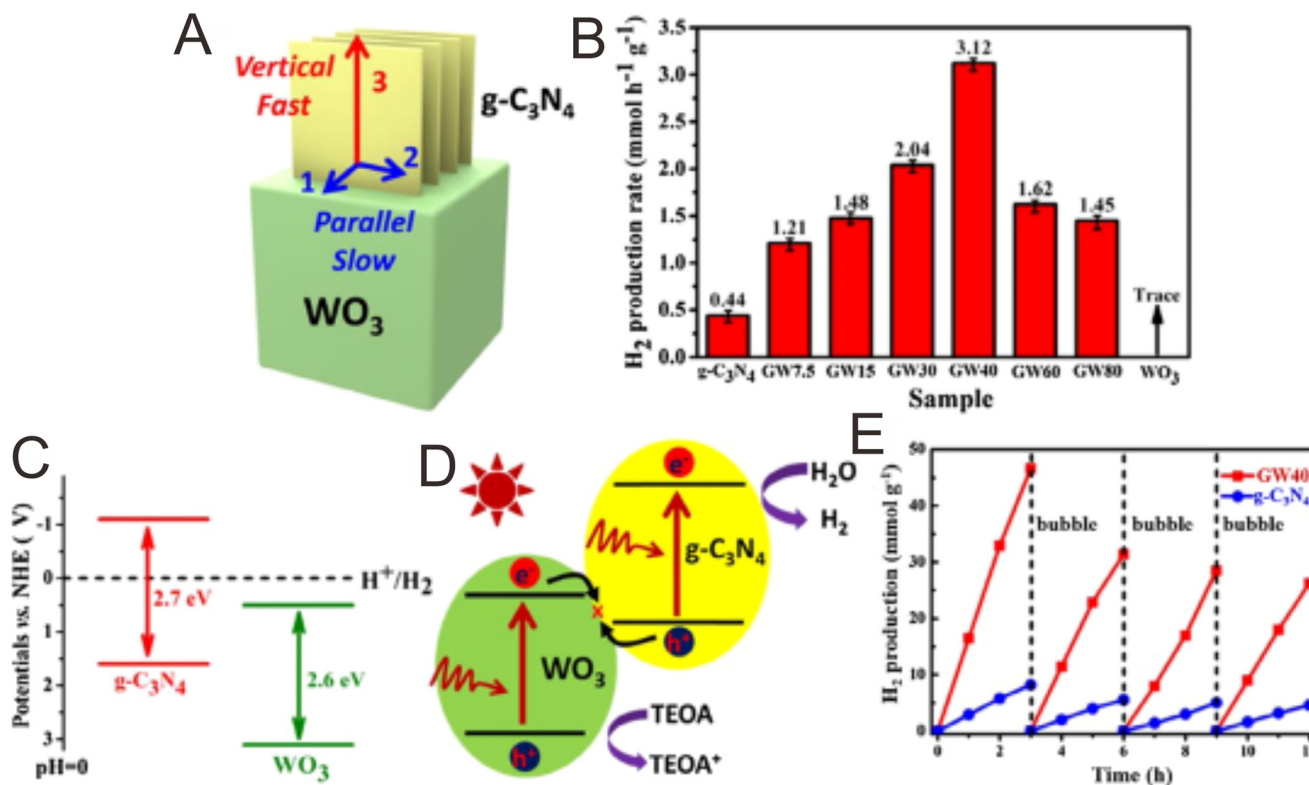
It is generally known that not all photogenerated charges centered on the photocatalysts' surface are utilized for the photocatalytic reaction. Typically, there are only some photogenerated e<sup>-</sup> in a semiconductor with enough redox potential that can be utilized for photocatalytic reactions.<sup>76,125,126</sup> 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 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<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).<sup>127</sup> If it follows type II mechanism, the photogenerated e<sup>-</sup> 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<sup>128</sup> proved the successful fabrication of the g-C<sub>3</sub>N<sub>4</sub>/CoTiO<sub>3</sub> direct Z-scheme heterojunctions through analyzing the 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<sup>-</sup> would be focused on CoTiO<sub>3</sub> 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

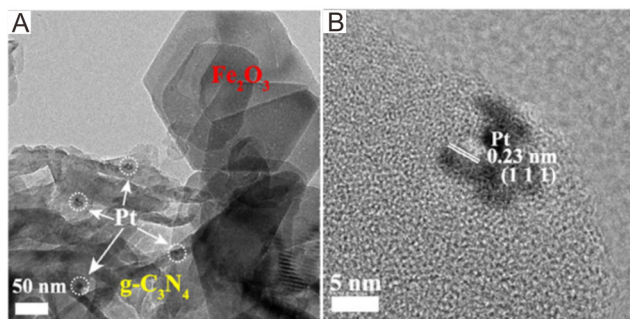
As for Z-scheme systems, the photogenerated charges can be efficiently separated. Photodeposition of several noble metals, *e.g.*, Au, Ag, Cu, on electron-rich areas is a promising strategy to unlock the photocatalytic reaction



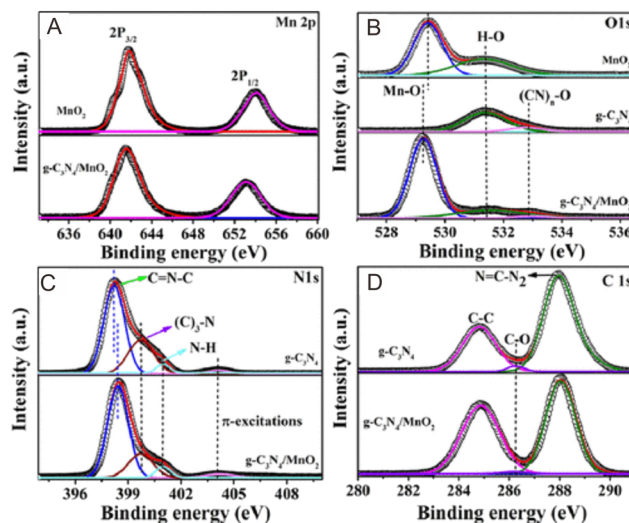
**Fig. 3** (A) The growth and extension of g-C<sub>3</sub>N<sub>4</sub> parallel with and perpendicular to the flat surface of the WO<sub>3</sub> nanocuboids. (B) Comparison of the photocatalytic H<sub>2</sub> production properties. (C) Band structure of g-C<sub>3</sub>N<sub>4</sub> and WO<sub>3</sub>. (D) The direct Z-scheme charge migration path in the g-C<sub>3</sub>N<sub>4</sub>/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.<sup>129</sup> 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<sup>-</sup> 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>/g-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. 137.

### 3.3. XPS characterization

XPS is a promising technique for analyzing the photocatalysts' surface chemistry,<sup>9,131–135</sup> 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.<sup>98,136</sup> 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  $g\text{-C}_3\text{N}_4/\text{MnO}_2$ ,<sup>137</sup> the Mn 2p spectrum of the  $\text{MnO}_2$  nanosheets (Fig. 5A) displayed two main peaks located at 641.9 and 654.0 eV, corresponding to Mn 2P<sub>3/2</sub> and Mn 2P<sub>1/2</sub>, 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)<sub>3</sub>-N), and N-H bonding in the  $g\text{-C}_3\text{N}_4$  framework, respectively. When compared with the  $\text{MnO}_2$  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  $\text{MnO}_2$  onto the  $g\text{-C}_3\text{N}_4$  nanosheets with electrons flowing from  $g\text{-C}_3\text{N}_4$  to  $\text{MnO}_2$  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.<sup>138</sup>

### 3.4. SPV techniques

Recently, the SPV strategy has provided some new opportunities to study the charge transfer path of Z-scheme systems.<sup>139</sup> Typically, Kelvin probe force microscopy (KPFM) can easily obtain the photocatalysts' surface potential distribution with high spatial resolution and potential sensitivity.<sup>140–143</sup> Therefore, using KPFM-based SPV techniques to study the carrier transfer path of Z-scheme systems is an efficient technology. Guan and co-workers<sup>144</sup> reported a  $g\text{-C}_3\text{N}_4$ -based Z-scheme system through encapsulation of  $g\text{-C}_3\text{N}_4$  into hollow  $\text{In}_2\text{O}_3$  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  $\text{In}_2\text{O}_3$ , further illustrating the Z-scheme mechanism. Therefore, the SPV strategy is also very promising for the confirmation of the Z-scheme mechanism.

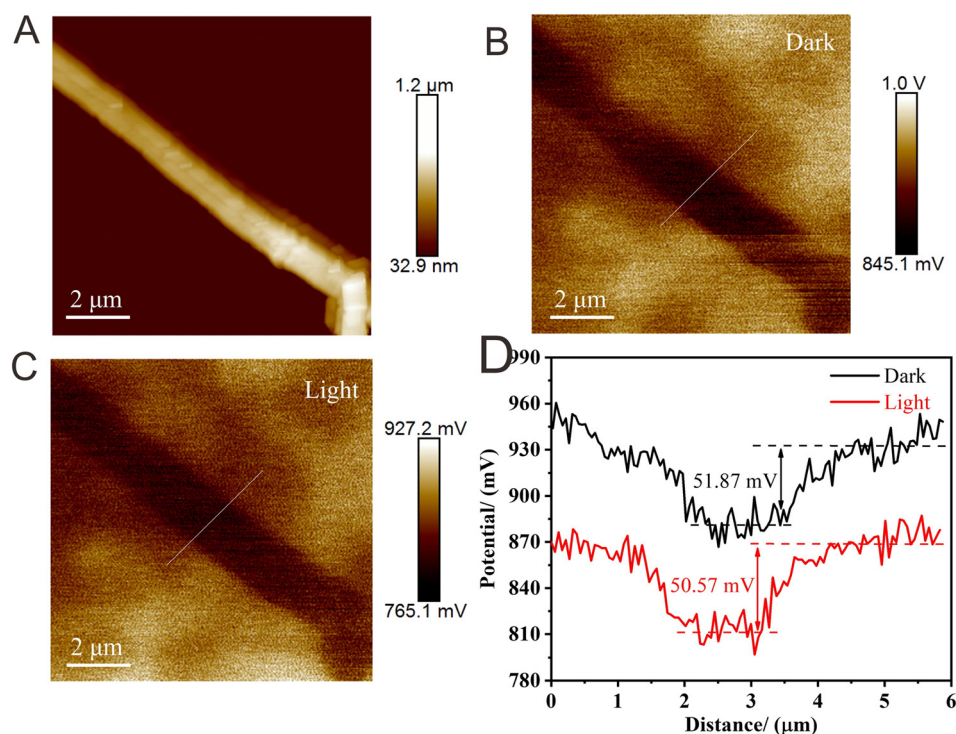


Fig. 6 (A) Atomic force microscopy image of the samples. (B) KPFM potential image of the samples under dark conditions. (C) KPFM potential image of the samples under light illumination. (D) The corresponding surface curves correspond to the white light. Reproduced with permission from ref. 144.

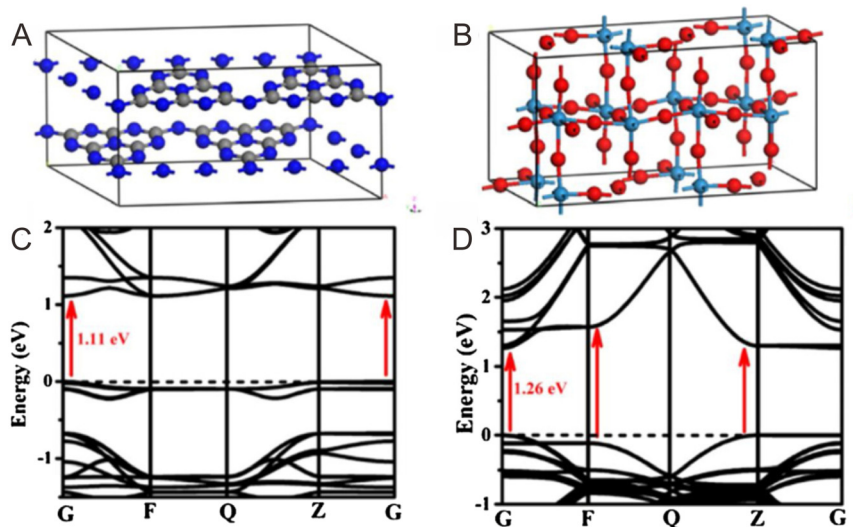


Fig. 7 Crystal structures of (A) g-C<sub>3</sub>N<sub>4</sub> and (B) monoclinic WO<sub>3</sub>. The calculated electronic band structures of (C) g-C<sub>3</sub>N<sub>4</sub> and (D) WO<sub>3</sub>. Reproduced with permission from ref. 127.

### 3.5. DFT calculation

It is well known that DFT calculation is a promising technology to explore the charge migration mechanism.<sup>145,146</sup> 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.<sup>108,127,147</sup> The DFT calculation is conducted based on the crystal and electronic band structures (Fig. 7).<sup>127</sup> The effective masses of the charges of WO<sub>3</sub> and g-C<sub>3</sub>N<sub>4</sub> 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):

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

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

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

where  $\hbar$ ,  $d^2E/dk^2$ ,  $k$ ,  $m^*$ ,  $v$ ,  $D$ ,  $m_h^*$ , 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-C<sub>3</sub>N<sub>4</sub> and WO<sub>3</sub>

| Species                         | Effective mass | G-F direction | G-Z direction | F-Q direction | Z-Q direction |
|---------------------------------|----------------|---------------|---------------|---------------|---------------|
| g-C <sub>3</sub> N <sub>4</sub> | $m_h^*$        | 0.72          | 71            | —             | —             |
|                                 | $m_e^*$        | 0.53          | 1.8           | —             | —             |
|                                 | $D$            | 1.36          | 39.44         | —             | —             |
| WO <sub>3</sub>                 | $m_h^*$        | 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<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<sup>-</sup> 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<sub>3</sub>N<sub>4</sub> ( $D = 1.36$ ) 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.<sup>127</sup>

## 4. Preparation methods for g-C<sub>3</sub>N<sub>4</sub>-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 for fabricating g-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts.<sup>148</sup> Narayanan and co-workers reported on a AgCl@g-C<sub>3</sub>N<sub>4</sub> direct Z-scheme photocatalyst through a modified deposition-precipitation method.<sup>149</sup> AgNO<sub>3</sub> 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<sup>+</sup> 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 | (i) Facile, eco-friendly, and highly efficient<br>(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-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, and homogeneous size distribution.<sup>13,150–155</sup> 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.<sup>157,158</sup> 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 milling<sup>159</sup> and dissolving precursors in a solvent followed by drying,<sup>160</sup> 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 TiO<sub>2</sub> 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.

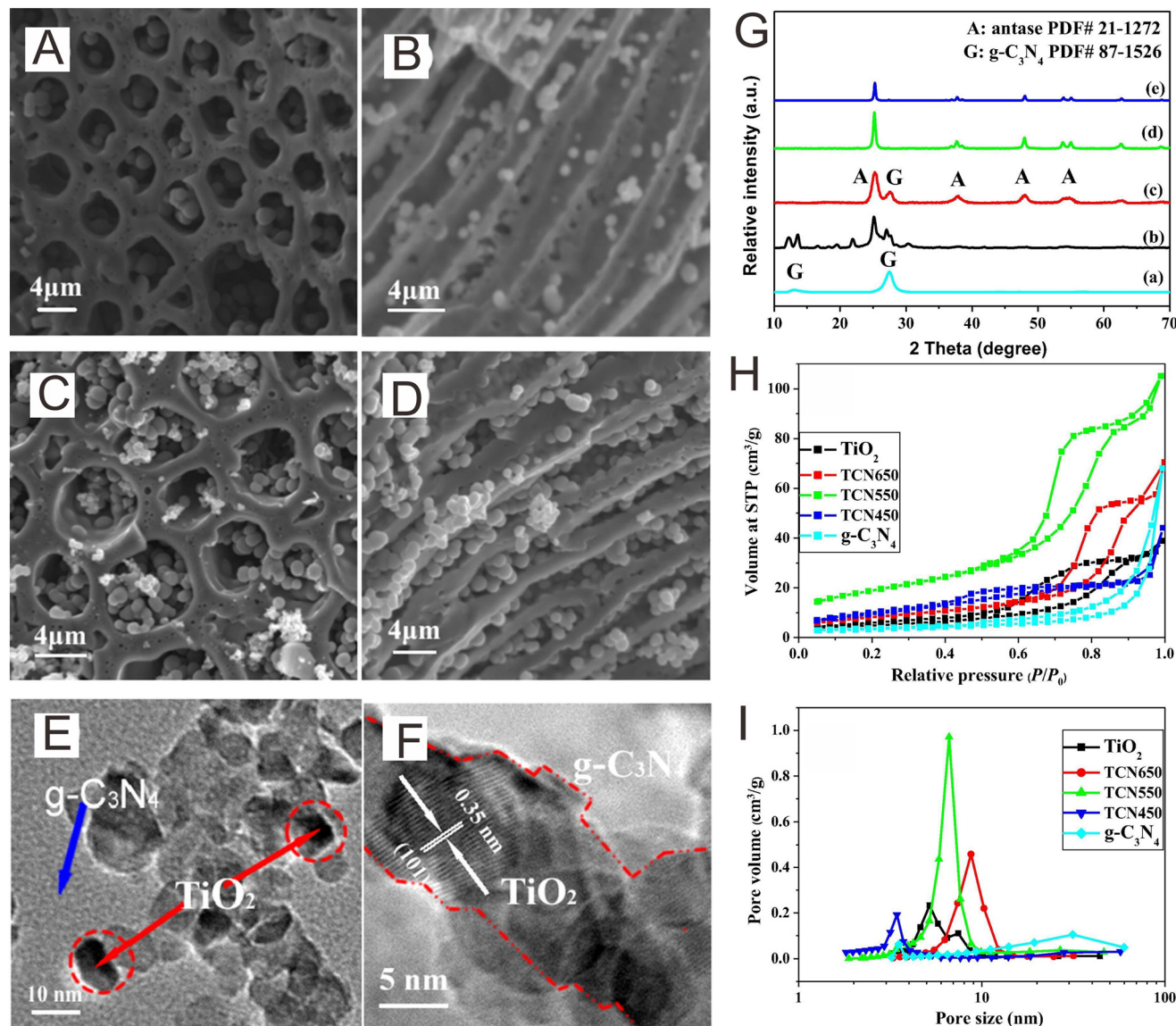
#### 4.4. Ion exchange strategy

The ion exchange method mainly involves exchanging ions in an ionic crystal, but the framework still keeps close intact with each other.<sup>162–164</sup> 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 H<sub>2</sub> 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.<sup>166,167</sup> 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.<sup>168,169</sup> Li and co-workers<sup>170</sup> 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 area,





**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), N<sub>2</sub> 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.<sup>8,171–174</sup> Cheng and co-workers<sup>175</sup> proposed a simple electrostatic self-assembly 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<sub>4</sub> 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.<sup>176</sup> 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.<sup>177–179</sup> 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.<sup>129</sup> The photo-deposition method is very

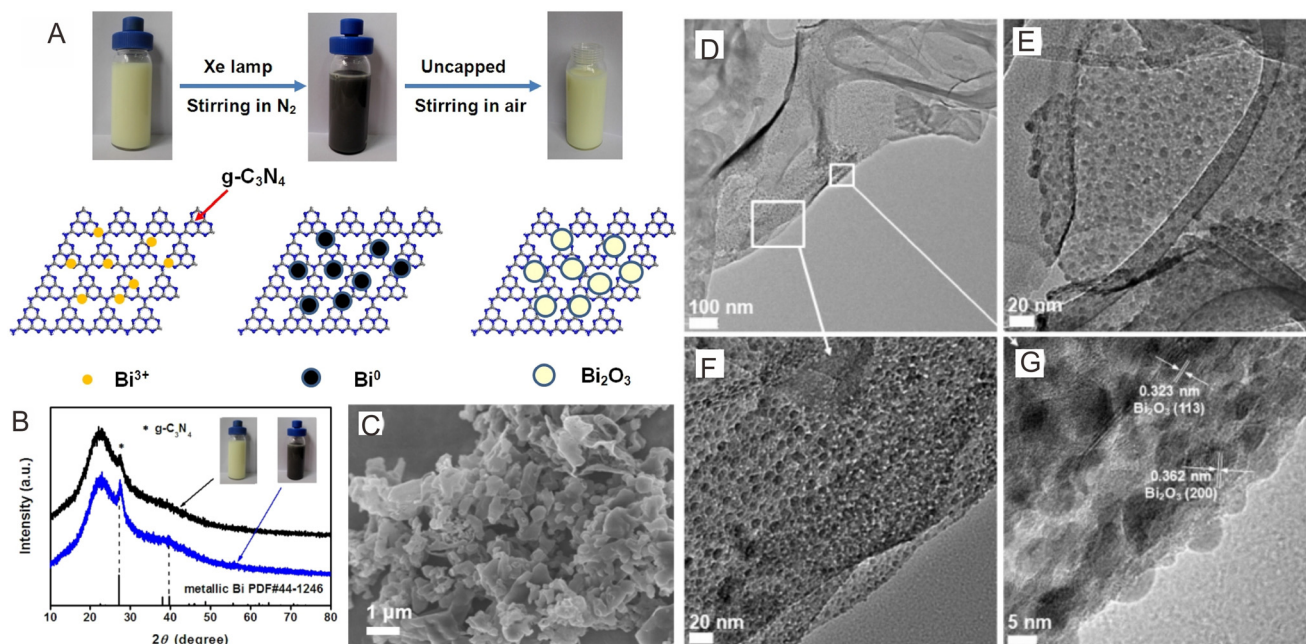


Fig. 9 (A) Diagrammatic sketch of the preparation of  $\text{Bi}_2\text{O}_3/\text{g-C}_3\text{N}_4$ . (B) XRD patterns of the intermediate products. (C) FESEM image of  $\text{Bi}_2\text{O}_3/\text{g-C}_3\text{N}_4$ . TEM (D–F) and HRTEM (G) images of  $\text{Bi}_2\text{O}_3/\text{g-C}_3\text{N}_4$ . 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  $\text{Bi}^{3+}$  on the  $\text{g-C}_3\text{N}_4$  surface under light illumination, followed by air oxidation of  $\text{Bi}^{3+}$  to  $\text{Bi}_2\text{O}_3$  (Fig. 9A).<sup>180</sup> The uniform dispersion of quantum-sized  $\text{Bi}_2\text{O}_3$  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-workers<sup>181</sup> 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  $\text{BiVO}_4$ , 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,<sup>182</sup> impregnation,<sup>183</sup> planetary ball-milling,<sup>184</sup> and reflux.<sup>185</sup> 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  $\text{g-C}_3\text{N}_4/\text{WO}_3$  Z-scheme photocatalysts *via* grinding, impregnation assisted with sonication, and planetary ball-milling methods. Different from the agglomerated  $\text{WO}_3$  decorated on the  $\text{g-C}_3\text{N}_4$  nanoplate prepared through the first two methods,  $\text{WO}_3$  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.<sup>76,188</sup> 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.<sup>189,190</sup> The self-assembly method can ensure the two semiconductors with a narrow size distribution and uniform morphology.<sup>191</sup> In contrast, the mechanical agitation method exhibits relatively poor interface contact between the two semiconductors, resulting in low photocatalytic performance.<sup>192</sup> 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

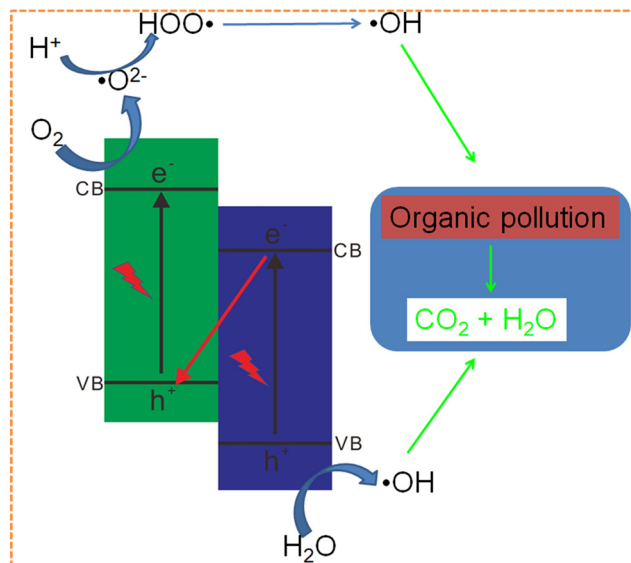


Fig. 10 Diagrammatic explanation of organic pollutant degradation over  $g\text{-C}_3\text{N}_4$ -based Z-scheme photocatalysts.

have recently been excessively discharged to soil and water, which have negative influences on the environment and human health.<sup>193–201</sup>  $g\text{-C}_3\text{N}_4$ -based Z-scheme photocatalysts have gained growing attention for organic pollutant removal because of their enhanced photocatalytic efficiency and stability (Fig. 10).<sup>77,101,202</sup> 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

$\text{TiO}_2$  is valuable in the photocatalytic fields because of its unique advantages, *e.g.*, excellent controllability, large surface area, amazing stability, and ease of recycling.<sup>203,204</sup> Some investigations have focused on combining  $\text{TiO}_2$  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,<sup>73</sup> who achieved markedly enhanced photocatalytic properties by  $g\text{-C}_3\text{N}_4/\text{TiO}_2$  Z-scheme photocatalysts. Dong and co-workers<sup>205</sup> fabricated an all-solid-state (ASS) Z-scheme photocatalyst through decorating  $g\text{-C}_3\text{N}_4$  nanosheets on  $\text{TiO}_2$  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  $\text{TiO}_2$  TNTAs. Ho and co-workers<sup>206</sup> prepared the  $g\text{-C}_3\text{N}_4$  quantum dots (QDs)-modified rutile  $\text{TiO}_2$  ( $r\text{TiO}_2$ ) ( $g\text{-C}_3\text{N}_4$  QDs- $r\text{TiO}_2$ ) hybrid *via* calcination of the mixture of P25  $\text{TiO}_2$  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  $\text{BiVO}_4$  have been demonstrated to be the active reduction and oxidation sites, respectively.<sup>207</sup> Raj and co-workers<sup>208</sup> 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 group<sup>209</sup> studied the influence of the exposed  $\{001\}$  and  $\{101\}$  facets of  $\text{TiO}_2$  on the photocatalytic activity. They first fabricated  $\text{TiO}_2$  nanosheets (NSs) and  $\text{TiO}_2$  nanoboxes (NBs), and then combined them with  $g\text{-C}_3\text{N}_4$  to prepare  $\text{TiO}_2$  NS/ $g\text{-C}_3\text{N}_4$  and  $\text{TiO}_2$  NBs/ $g\text{-C}_3\text{N}_4$ , respectively (Fig. 11A and B). The  $\{001\}$  facets of  $\text{TiO}_2$  NSs came into contact with  $g\text{-C}_3\text{N}_4$  (Fig. 11A and C), and the  $\{101\}$  facets of  $\text{TiO}_2$  NBs came into contact with  $g\text{-C}_3\text{N}_4$  (Fig. 11B and D). Under visible light illumination, the photogenerated carriers on  $\text{TiO}_2$  were separately accumulated on the  $\{101\}$  and  $\{001\}$  facets.<sup>210</sup> The photogenerated electrons were subsequently centered on the  $\{101\}$  facets of the  $\text{TiO}_2$  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  $\text{TiO}_2$  NBs/ $g\text{-C}_3\text{N}_4$  is much higher than that of  $\text{TiO}_2$  NSs/ $g\text{-C}_3\text{N}_4$ , and the  $\text{TiO}_2$  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

$\text{WO}_3$  is gradually becoming recognized as one of the most promising photocatalysts, owing to its outstanding photocatalytic performance, chemical inertness, and optical stability.<sup>211,212</sup> In comparison with  $\text{TiO}_2$ ,  $\text{WO}_3$  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 group<sup>213</sup> 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 group<sup>214</sup> 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  $\text{WO}_3$ , respectively. Kang and co-workers<sup>215</sup> 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-workers<sup>216</sup> 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-workers<sup>217</sup> 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-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts

| Photocatalysts   | Preparation strategy                             | Mass (mg) | Light source                   | Target pollutant/initial concentration   | Degradation time (min) and rate (%) | $K_{app}^a$ [ $10^{-2} \text{ min}^{-1}$ ] | 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 mg L <sup>-1</sup>   | 120 (~97)                           | 0.95                                       | 208  |
| g-C <sub>3</sub> N <sub>4</sub> /TiO <sub>2</sub> -HNB                             | Microwave-assisted strategy                      | —         | 3 W LED lamp, 365 nm           | Brilliant red X-3B/1.0 × 10 <sup>-4</sup> mmol L <sup>-1</sup>                   | 45 (~99)                            | 5.1  | 209  |
| WO <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>                                   | Ball milling and heat treatment methods          | 10        | 500 W xenon lamp, >470 nm      | MB and BF/0.9 × 10 <sup>-5</sup> and 1.0 × 10 <sup>-5</sup> mol L <sup>-1</sup>  | MB 120 (~99) BF 120 (~97)           | —  | 213  |
| WO <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>                                   | <i>In situ</i> liquid phase process              | 50        | 300 W xenon lamp, >400 nm      | MB/50 mg L <sup>-1</sup>   | 90 (~97)                            | 1.90                                       | 214  |
| WO <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>                                   | 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 strategy                      | 100       | 500 W xenon lamp, >420 nm      | TC/10 mg L <sup>-1</sup>   | 60 (~70)                            | —  | 217  |
| WO <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub> /Bi <sub>2</sub> O <sub>3</sub>   | One step co-calcination way                      | 100       | 300 W xenon lamp, >420 nm      | TC/10 mg L <sup>-1</sup>   | 60 (~80)                            | 2.37                                       | 159  |
| g-C <sub>3</sub> N <sub>4</sub> /MnO <sub>2</sub>                                  | Wet-chemical method                              | 50        | 300 W xenon lamp               | RhB/10 mg L <sup>-1</sup>  | 180 (~77)                           | 3.3  | 137  |
| Bi <sub>2</sub> O <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>                    | Ball milling and heat treatment methods          | 5         | 500 W xenon lamp, 470 nm       | MB and RhB/1.1 × 10 <sup>-5</sup> and 1.0 × 10 <sup>-5</sup> mol L <sup>-1</sup> | MB 120 (~76) RhB 120 (~73)          | 2.53 and 1.01                              | 223  |
| 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 mg L <sup>-1</sup>   | 160 (~88)                           | 1.15                                       | 225  |
| β-Bi <sub>2</sub> O <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>                  | Hydrothermal calcination approach                | 40        | 350 W xenon lamp, >420 nm      | RhB/1 × 10 <sup>-5</sup> mol/L   | 80 (~97)                            | 4.22                                       | 226  |
| g-C <sub>3</sub> N <sub>4</sub> /Bi <sub>4</sub> O <sub>7</sub>                    | 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 <sup>-1</sup>  | 120 (>99)                           | 1.13                                       | 228  |
| g-C <sub>3</sub> N <sub>4</sub> /Bi <sub>2</sub> WO <sub>6</sub>                   | One-pot hydrothermal strategy                    | 50        | Xenon lamp (XL-300, Yirda)     | RhB/10 mg L <sup>-1</sup>  | 30 (>99)                            | 8.17                                       | 156  |
| g-C <sub>3</sub> N <sub>4</sub> /RGO/Bi <sub>2</sub> WO <sub>6</sub>               | Hydrothermal method                              | 250       | 500 W xenon lamp, >420 nm      | TCP/20–100 mg L <sup>-1</sup>  | 120 (>99)                           | 3.15                                       | 229  |
| Ag-decorated g-C <sub>3</sub> N <sub>4</sub> /Bi <sub>2</sub> WO <sub>6</sub>      | Polymerization method                            | 40        | 300 W xenon lamp, 400 nm       | RhB/10 mg L <sup>-1</sup>  | 90 (>99)                            | —  | 230  |
| Bi <sub>3</sub> O <sub>4</sub> Cl/g-C <sub>3</sub> N <sub>4</sub>                  | Solid-phase calcination method                   | 50        | 250 W xenon lamp, >420 nm      | RhB and TC/10 and 10 mg L <sup>-1</sup>  | RhB 60 (>98) TC 60 (>76)            | — and 2.05                                 | 231  |
| BiOBr/RGO/pg-C <sub>3</sub> N <sub>4</sub>   | Solvothermal method                              | 50        | 300 W xenon lamp, 420 nm       | RhB and TC/10 and 10 mg L <sup>-1</sup>  | RhB 7.5 (>99) TC 60 (>97)           | — and —                                    | 232  |
| Bi <sub>12</sub> GeO <sub>20</sub> /g-C <sub>3</sub> N <sub>4</sub>                | Sol-gel method                                   | 300       | 300 W Dy lamp, 400 nm          | RhB/5 mg L <sup>-1</sup>   | 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 method                   | 100       | 500 W xenon lamp, >420 nm      | β-Naphthol/15 mg L <sup>-1</sup>   | 150 (>62)                           | 0.63                                       | 234  |
| Ag <sub>3</sub> PO <sub>4</sub> /g-C <sub>3</sub> N <sub>4</sub>                   | <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 <sub>3</sub> PO <sub>4</sub> /g-C <sub>3</sub> N <sub>4</sub>                   | <i>In situ</i> precipitation method              | 30        | 300 W xenon lamp               | MO/10 mg L <sup>-1</sup>   | 5 (>99)                             | —  | 239  |
| g-C <sub>3</sub> N <sub>4</sub> /Ag <sub>2</sub> WO                                | <i>In situ</i> precipitation method              | 30        | 300 W xenon lamp               | MO/10 mg L <sup>-1</sup>   | 150 (~98)                           | —  | 240  |
| Ag <sub>2</sub> CrO <sub>4</sub> /g-C <sub>3</sub> N <sub>4</sub>                  | Precipitation method                             | 20        | 300 W xenon lamp, >400 nm      | MO and RhB/10 and 10 mg L <sup>-1</sup>  | MO 21 (~90) RhB 40 (~97)            | 9.64 and 9.21                              | 241  |
| Ag@AgBr/g-C <sub>3</sub> N <sub>4</sub>  | Deposition-precipitation method                  | 100       | 300 W xenon lamp               | MO and MB/10 and 10 mg L <sup>-1</sup>   | MO 10 (~97) RhB 10 (~96)            | — and —                                    | 117  |
| Ag <sub>2</sub> CrO <sub>4</sub> /Ag/g-C <sub>3</sub> N <sub>4</sub>               | <i>In situ</i> growth and photoreduction methods | 50        | 500 W xenon lamp, <420 nm      | 2,4-DCP/10 mg L <sup>-1</sup>  | 120 (~96)                           | 1.52                                       | 243  |
| g-C <sub>3</sub> N <sub>4</sub> /Ag/MoS <sub>2</sub>                               | <i>In situ</i> 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 <sup>-4</sup> mol L <sup>-1</sup>  | 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 strategy                    | 50        | 300 W xenon lamp, >420 nm      | RhB/30 mg L <sup>-1</sup>  | 180 (~92)                           | 1.37                                       | 250  |
| MoO <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>                                  | Calcination method                               | 30        | 350 W xenon lamp, >420 nm      | TC/20 mg L <sup>-1</sup>   | 90 (~90)                            | 2.23                                       | 251  |
| p-type Cu <sub>3</sub> P/n-type g-C <sub>3</sub> N <sub>4</sub>                    | 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 (%) | $K_{app}^a$ [ $10^{-2} \text{ min}^{-1}$ ] | 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 <sub>2</sub> O <sub>5</sub> /g-C <sub>3</sub> N <sub>4</sub>                       | <i>In situ</i> growth strategy     | 50        | 250 W xenon lamp, 420 nm  | RhB and TC/10 and 10 mg L <sup>-1</sup> | 120 (~77)                           | 4.91 and —                                 | 107  |
| SnO <sub>2</sub> /g-C <sub>3</sub> N <sub>4</sub>                                    | Calcination method                 | 100       | 350 W xenon lamp, >420 nm | RhB/10 mg L <sup>-1</sup>               | 40 (>99)                            | 1  | 254  |
| InVO <sub>4</sub> /Bi <sub>2</sub> S <sub>3</sub> /g-C <sub>3</sub> N <sub>4</sub>   | 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 <sub>3</sub> N <sub>4</sub> /SnO <sub>2</sub>                                | Thermal polymerization             | 10        | 300 W xenon lamp, >420 nm | IDM/10 mg L <sup>-1</sup>               | 40 (>99)                            | 2.33                                       | 256  |
| g-C <sub>3</sub> N <sub>4</sub> /ZnFe <sub>2</sub> O <sub>4</sub> /UiO-66            | Hydrothermal method                | —         | 300 W xenon lamp, >420 nm | RhB/—                                   | 40 (>99)                            | 4.89                                       | 256  |
| g-C <sub>3</sub> N <sub>4</sub> /BiVO <sub>4</sub> /CoFe <sub>2</sub> O <sub>4</sub> | High-temperature solid-state route | 50        | 300 W xenon lamp, >400 nm | RhB/—                                   | 25 (>99)                            | 2.84                                       | 257  |
| g-C <sub>3</sub> N <sub>4</sub> /g-C <sub>3</sub> N <sub>4</sub>                     | Bottom-up approach                 | 10        | 50 W xenon lamp, >400 nm  | RhB/10 mg L <sup>-1</sup>               | 240 (>99)                           | 1.39                                       | 246  |

<sup>a</sup>  $K_{app}$  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 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.

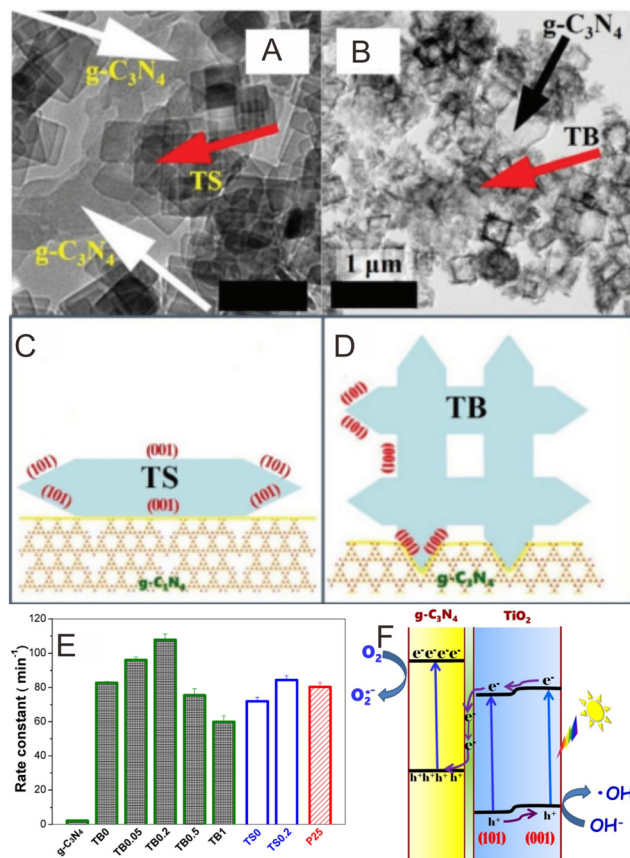
### 5.3. g-C<sub>3</sub>N<sub>4</sub>/MnO<sub>2</sub> Z-scheme photocatalysts

MnO<sub>2</sub> 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).<sup>219,220</sup> 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-C<sub>3</sub>N<sub>4</sub> and MnO<sub>2</sub> (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,<sup>221,222</sup> and the CB of g-C<sub>3</sub>N<sub>4</sub> and MnO<sub>2</sub> 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<sup>-</sup> in MnO<sub>2</sub> cannot generate ·O<sup>2-</sup> radicals, respectively, because of the thermodynamic equilibrium limit. After the Z-scheme heterojunction was formed, the photogenerated e<sup>-</sup> 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<sub>2</sub>O<sub>3</sub>,<sup>159,180,223–225</sup> β-Bi<sub>2</sub>O<sub>3</sub>,<sup>226</sup> Bi<sub>4</sub>O<sub>7</sub>,<sup>227</sup> Bi<sub>2</sub>WO<sub>6</sub>,<sup>156,228–230</sup> Bi<sub>3</sub>O<sub>4</sub>Cl,<sup>231</sup> BiOBr,<sup>232</sup> Bi<sub>12</sub>GeO<sub>20</sub>,<sup>233</sup> Bi<sub>2</sub>MoO<sub>6</sub>,<sup>234</sup> 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, and highly efficient photocatalytic performance.<sup>235–237</sup> For example, Chen and co-workers<sup>223</sup> 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 than 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 TiO<sub>2</sub> and g-C<sub>3</sub>N<sub>4</sub> on (C) TiO<sub>2</sub> NSs/g-C<sub>3</sub>N<sub>4</sub> and (D) TiO<sub>2</sub> NBs/g-C<sub>3</sub>N<sub>4</sub>. (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 as-prepared 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 Bi<sub>2</sub>WO<sub>6</sub> by 7.6 and 8.9 times, respectively. Yin and co-workers<sup>233</sup> prepared a new ASS Bi<sub>12</sub>GeO<sub>20</sub>/g-C<sub>3</sub>N<sub>4</sub> 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 Bi<sub>12</sub>-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 g-

C<sub>3</sub>N<sub>4</sub> to fabricate Z-scheme photocatalysts for enhancing photocatalytic properties owing to their suitable CB and CB values.

### 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 Ag-containing semiconductors/g-C<sub>3</sub>N<sub>4</sub> SCS Z-scheme 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 *via* the 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 co-workers<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 a metal-free 2D g-C<sub>3</sub>N<sub>4</sub>/g-C<sub>3</sub>N<sub>4</sub> 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-C<sub>3</sub>N<sub>4</sub>/g-C<sub>3</sub>N<sub>4</sub> Z-scheme photocatalysts showed better 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.<sup>33</sup> 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 ~0.4–0.6 μm (Fig. 13A). The prepared g-C<sub>3</sub>N<sub>4</sub>/Ag/MoS<sub>2</sub> showed

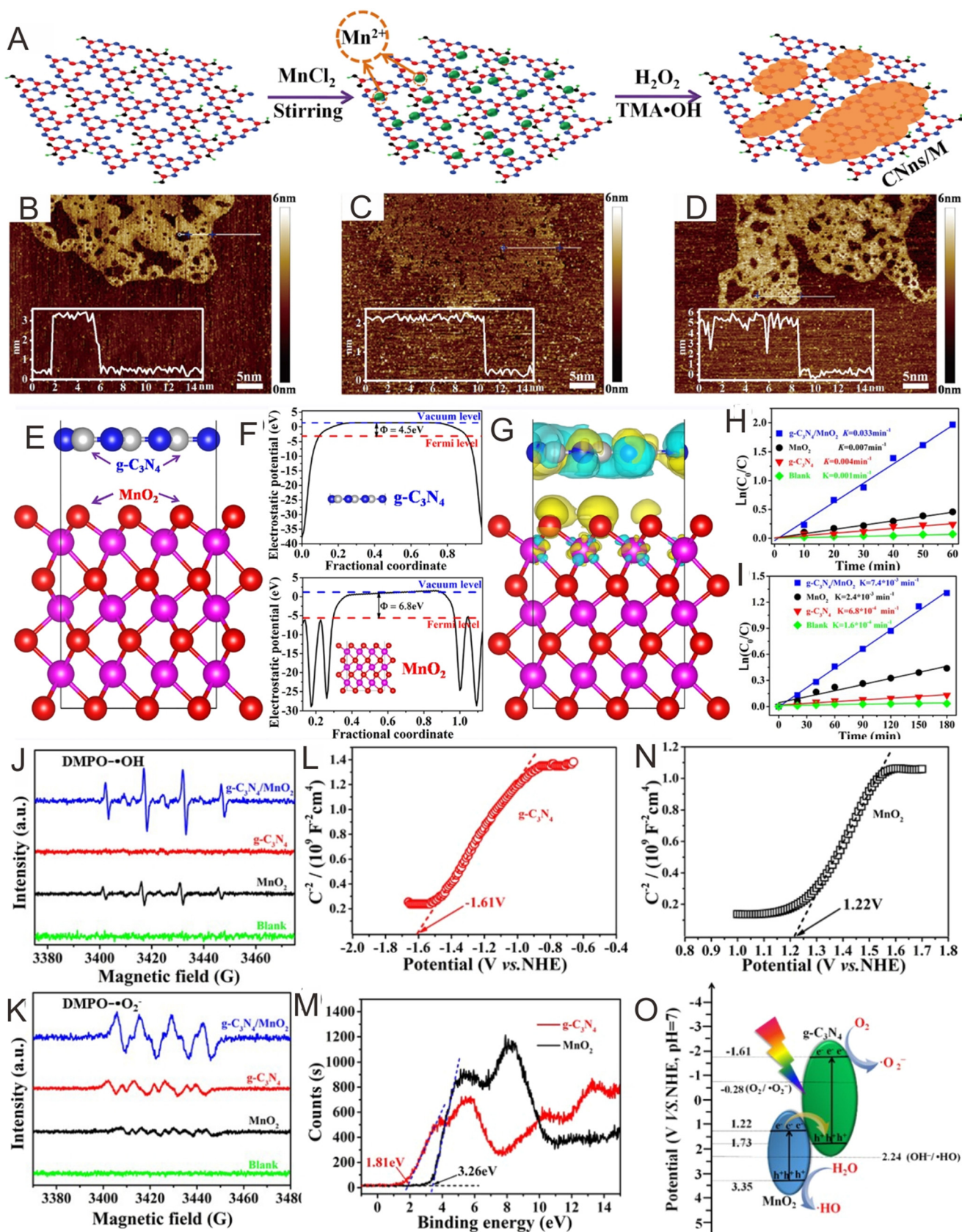


Fig. 12 (A) Diagrammatic sketch of  $g\text{-C}_3\text{N}_4/\text{MnO}_2$  prepared through *in situ* growth of  $\text{MnO}_2$  on the  $g\text{-C}_3\text{N}_4$  nanosheets. AFM images of the (B)  $g\text{-C}_3\text{N}_4$  nanosheets, (C)  $\text{MnO}_2$  nanosheets, and (D)  $g\text{-C}_3\text{N}_4/\text{MnO}_2$ . (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  $\text{MnO}_2$  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  $g\text{-C}_3\text{N}_4$ ,  $\text{MnO}_2$ , 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  $g\text{-C}_3\text{N}_4$ ,  $\text{MnO}_2$ , 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)  $\text{MnO}_2$  nanosheets. (N) XPS valence band spectra of  $\text{MnO}_2$  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.

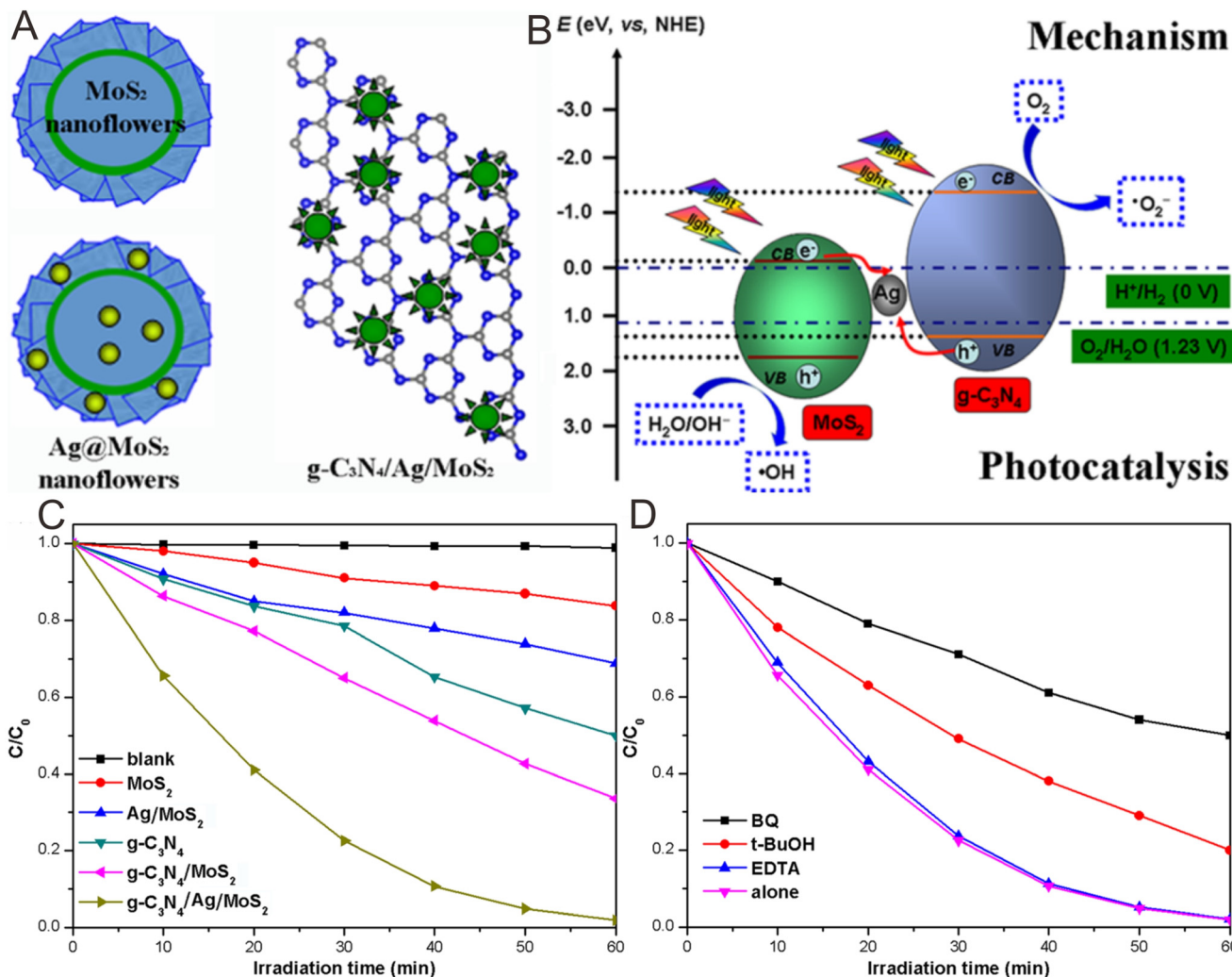


Fig. 13 (A) The nanostructures of Ag/MoS<sub>2</sub> and g-C<sub>3</sub>N<sub>4</sub>/Ag/MoS<sub>2</sub>. (B) Diagrammatic sketch for the separation and migration of the photogenerated charges over g-C<sub>3</sub>N<sub>4</sub>/Ag/MoS<sub>2</sub>. (C and D) Photocatalytic property of the fabricated MoS<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>/Ag/MoS<sub>2</sub> photocatalysts for RhB solution. Reproduced with permission from ref. 247.

significantly enhanced photocatalytic performance, exceeding that of the Ag/MoS<sub>2</sub> and g-C<sub>3</sub>N<sub>4</sub>/MoS<sub>2</sub> 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-C<sub>3</sub>N<sub>4</sub>, and MoS<sub>2</sub> nanophase structures, which led to a Z-scheme-mechanism-induced rapid separation of the photogenerated e<sup>-</sup>-h<sup>+</sup> pairs and further 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<sub>3</sub>N<sub>4</sub>-RGO-TiO<sub>2</sub>,<sup>250</sup> MoO<sub>3</sub>/g-C<sub>3</sub>N<sub>4</sub>,<sup>251</sup> Cu<sub>3</sub>P/g-C<sub>3</sub>N<sub>4</sub>,<sup>252</sup> 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> g-

C<sub>3</sub>N<sub>4</sub>/ZnFe<sub>2</sub>O<sub>4</sub>/UiO-66,<sup>256</sup> and g-C<sub>3</sub>N<sub>4</sub>/BiVO<sub>4</sub>/CoFe<sub>2</sub>O<sub>4</sub>.<sup>257</sup> 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-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.<sup>260</sup> 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-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<sub>3</sub>N<sub>4</sub>-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-C<sub>3</sub>N<sub>4</sub>-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-C<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts is a promising method for improving their photocatalytic performance.

(4) The photocatalytic performance of g-C<sub>3</sub>N<sub>4</sub>-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<sub>3</sub>N<sub>4</sub>-based Z-scheme photocatalysts that will be very good for further enhancing its e<sup>-</sup>-h<sup>+</sup> separation and light utilization.

(5) Considering the unique advantages of g-C<sub>3</sub>N<sub>4</sub>-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<sub>3</sub>N<sub>4</sub>-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.

## Acknowledgements

This work was supported by the National Natural Science Foundation of China (Grant No. 52203110), the Knowledge Innovation Program of Wuhan-Shuguang Project (Grant No. 2022010801020216), the Scientific Research Foundation of Fujian University of Technology (Grant No. GY-Z220182), the China Postdoctoral Science Foundation (Grant No. 2021M693795), and the Excellent Youth Foundation of Jiangsu Science Committee (Grant No. BK20220118).

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