SPECIAL TOPIC: Advanced Photocatalytic Materials

2D/2D heterostructured photocatalyst: Rational design for energy and environmental applications

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ABSTRACT Two-dimensional/two-dimensional (2D/2D) hybrid nanomaterials have triggered extensive research in the photocatalytic field. The construction of emerging 2D/2D heterostructures can generate many intriguing advantages in exploring high-performance photocatalysts, mainly including preferable dimensionality design allowing large contact interface area, integrated merits of each 2D component and rapid charge separation by the heterojunction effect. Herein, we provide a comprehensive review of the recent progress on the fundamental aspects, general synthesis strategies (*in situ* growth and *ex situ* assembly) of 2D/2D heterostructured photocatalysts and highlight their applications in the fields of hydrogen evolution, CO₂ reduction and removal of pollutants. Furthermore, the perspectives on the remaining challenges and future opportunities regarding the development of 2D/2D heterostructure photocatalysts are also presented.

Keywords: 2D/2D heterojunction, photocatalyst, hydrogen evolution, CO₂ reduction, pollutant removal

INTRODUCTION

Two-dimensional (2D) materials have been deemed as rapidly rising stars in materials science, because they display a variety of extraordinary physicochemical and mechanical properties different from their bulk counterparts, such as large surface-to-volume ratio, high transparency and good flexibility [1–5]. Since the groundbreaking discovery of mono-layered graphene [6], plenty of newly developed 2D materials have been applied in many fields including energy storage [7], electrocatalysis [8], sensing [9], and photocatalysis [10]. Particularly, the utilization of 2D materials for photocatalysis has triggered considerable interest, and promptly become one of the hottest research topics [11–13]. It has been

demonstrated that 2D material-based photocatalysts are expected to offer intriguing features such as high specific surface areas, porous structures, good conductivity and superior electron mobility, rich options of host-guest species and abundant surface active sites, which are beneficial to photocatalytic reactions [14,15]. Moreover, heterostructures of 2D materials further offer an effective way for enhancing light absorption, and promoting the separation and transfer of photogenerated charge carriers [16–19]. Typically, it is well-known that the appropriate band alignments thermodynamically between the components should be firstly required in heterostructure design. By coupling the different band structures in various materials, the light responsive range is broadened while the charge separation and transportation is synchronously promoted, leading to enhanced photocatalytic activities [20–23]. However, besides the band structure matching, the interface contact, governed by the dimensionality of each component, is also critically vital for the smooth transfer of the photogenerated charge carriers. In regard to the dimensionality, 2D based heterostructures composed of two components can be classified into 0D/2D, 1D/2D and 2D/2D heterostructures with three different types of interfacial contacts as illustrated in Fig. 1. Distinctly, the face-to-face stacking of 2D/2D heterostructures has the largest interfacial contact area and strongest interactions, which enables relatively better coupling hetero-interfaces compared with 0D/2D and 1D/2D heterostructures [10,15]. The large contact area and strong interactions can also facilitate the transfer and separation of photogenerated electron-hole pairs, thus contributing to high photocatalytic performance. Profiting from these features, 2D/2D photocatalyst can display enormous prospects and a large number of related studies

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Figure 1 Schematic illustration of 2D based heterostructures in regard to the dimensionality difference.

have been reported in recent years. Although the research progress of 2D/2D heterostructure photocatalysts has been mentioned in some excellent reviews on 2D based photocatalysts [10,11,24,25], a thoroughly comprehensive review to specifically highlight this subject has yet been reported. Thus, it is highly desirable and urgent to present a comprehensive review on this subject to provide a better understanding of the state-of-the-art progress in this research field as well as promote the further development of 2D/2D heterostructured photocatalysts.

Here, we present a comprehensive review to summarize the advanced progress of 2D/2D heterostructure photocatalysts (Fig. 2). It begins with a brief discussion of the fundamental aspects of 2D/2D photocatalysts. Then, the emerging strategies for designing various 2D/2D heterostructures are briefly summarized. Subsequently, we highlight the recent progress of 2D/2D heterostructures for photocatalytic applications, including photocatalytic hydrogen production, carbon dioxide (CO_2) reduction and the removal of pollutions. Finally, a brief summary of the current the research status and key issues, along with the perspectives of 2D/2D heterostructures for photocatalysis applications will also be discussed.

FUNDAMENTAL ASPECTS OF 2D/2D HETEROJUNCTION PHOTOCATALYSTS

The interfacial coupling in 2D/2D photocatalysts

The activity and stability of the 2D/2D photocatalysts are highly dependent on the type and quality of the interface in 2D/2D photocatalysts [26]. Generally, the layers in 2D material heterostructures can be combined by covalent bonding and van der Waals (vdW) forces [27]. To form a well-defined covalent bonding interface in 2D/2D photocatalysts, it needs not only the lattice constant matching, but also valence matching on each side of the interface [3,27] (Fig. 3a). In contrast to the covalent bonding interfaces, the vdW forces between the layers in 2D material heterostructures are relatively weak. In addition, the vdW integration does not rely on one-to-one lattice-matching and valence-matching, ultimately en-

Figure 2 Schematic diagram for the scope of this review.

Figure 3 (a) Schematic illustrations of bonded heterostructure interface with a lattice-matched interface. (b) Bonding free atomic structure at a vdW interface. Reprinted with permission from Ref. [3]. Copyright 2019, Nature.

abling a broader heterostructure phase space (Fig. 3b). Although vdW force is weak, the formed interaction can still mediate various types of coupling across the 2D/2D interface [3,27]. For example, it can redistribute the charges at interfaces to balance the chemical potential, which would result in band bending phenomena in the 2D/2D photocatalysts. In recent years, the form of 2D/2D photocatalysts with vdW interface has attracted considerable interests because of its bond-free integration feature.

Characterization of interfaces in 2D/2D photocatalysts

Besides the interfacial coupling type, the characterization

of interface is also vital to guarantee its quality in 2D/2D photocatalysts [26]. To investigate the quality and interaction between the 2D components, many powerful characterization techniques have been employed, mainly including transmission electron microscopy (TEM), atomic force microscopy (AFM), X-ray photoelectron spectroscopy (XPS), and Kelvin probe force microscopy (KPFM) [26].

Low-magnification TEM is often used to explore the lateral size and roughly measure the thickness of the 2D nanosheet. However, the high-resolution TEM (HRTEM) is often performed to reveal the interfaces between the 2D/2D photocatalysts. AFM is an useful technique to accurately measure the thickness of 2D materials. When it is used to characterize the interface in 2D/2D materials, it should be switched to a conductive AFM (CAFM) mode, which is a variation of AFM. Then, the CAFM can provide a conductivity mapping to clearly explore the interface between two 2D materials. XPS is a well-known surface-sensitive and spectroscopic technique to identify the elemental composition of a material. Specially, the formation of the interface would cause a change in the electron configuration owing to the interaction between the 2D materials in 2D/2D photocatalysts. In this case, XPS can be employed to examine the interaction intensity between the 2D materials. It has been confirmed that the interfacial interaction between the 2D materials could alter the surface potential of a catalyst. KPFM is considered as a powerful technique to study the surface potential and reveal the interfacial interaction between 2D materials.

SYNTHESIS METHODS FOR 2D/2D HETEROJUNCTION PHOTOCATALYSTS

In situ growth of 2D/2D heterojunction photocatalysts

The *in situ* growth of 2D/2D heterostructures is normally implemented through the following steps. One of the two 2D materials is firstly synthesized. Then, another 2D component directly grows on the primarily synthesized 2D substrate through one-step or multi-step conversion approaches, such as wet-chemical synthesis and chemical vapor deposition (CVD) methods. Generally, the wet-chemical strategy is one of the most commonly used methods for assembling 2D/2D heterostructures due to the simplicity, low cost and high tunability of composition, mainly including hydrothermal/solvothermal, solution deposition and ion exchange. A large variety of 2D/2D heterostructures have been synthesized by this route, which are briefly summarized in Table 1.

As for the hydrothermal/solvothermal method, the pre-synthesized 2D materials were immersed in the precursor solution of the second 2D component to obtain a mixed suspension. The 2D/2D heterostructures were produced by the hydrothermal or solvothermal treatment of the suspension. For instance, Cao *et al.* [28] developed a $\text{Ti}_3\text{C}_2/\text{Bi}_2\text{WO}_6$ 2D/2D heterostructure by an *in situ* hydrothermal growth of Bi_2WO_6 nanosheets on ultrathin Ti_3C_2 nanosheets. As shown in Fig. 4a, few-layers ultrathin Ti_3C_2 nanosheets were firstly obtained by etching bulk Ti_3AlC_2 combining with ultrasonic exfoliation process. Because of the negative potential of Ti_3C_2 , Bi^{3+} cations could be adsorbed on the Ti_3C_2 nanosheet surface through electrostatic attraction in the $\text{Bi}(\text{NO}_3)_3/\text{Ti}_3\text{C}_2$ suspension. Subsequently, Na_2WO_6 -cetyl trimethyl ammonium bromide (CTAB) mixed solution was added into the above suspension. Finally, $\text{Ti}_3\text{C}_2/\text{Bi}_2\text{WO}_6$ 2D/2D heterostructure was formed after a hydrothermal treatment. In some other studies, the second 2D component was amorphous after hydrothermal reaction and thus an additional calcination process was often required to crystallize it. For example, Cheng and coworkers [29] firstly obtained C_3N_4 nanosheets by ultrasonic exfoliation of bulk C_3N_4 , and then immersed them in $\text{Bi}(\text{NO}_3)_3/\text{H}_2\text{O}/\text{Ti}(\text{OC}_3\text{H}_7)_4/\text{NaBH}_4$ benzyl alcohol solution. The 2D/2D $\text{C}_3\text{N}_4/\text{Bi}_{20}\text{TiO}_{32}$ heterojunctions were obtained by hydrothermal reaction and following calcination (Fig. 4b).

Another wet-chemical method for *in situ* growth of the second 2D component is solution deposition. Similarly, the first 2D nanosheet is pre-synthesized and then dispersed into some specific solvents to obtain suspension. In this process, one or more reactant ions are firstly adsorbed on the surface of the pre-synthesized 2D substrate through electrostatic attraction. Then, other reaction solutions are added into the mixed suspension under stirring and kept for some times to generate the second 2D component through chemical precipitation reaction. Compared with the hydrothermal or solvothermal process, the whole process of this route is at room temperature and atmospheric pressure, but only several specific 2D/2D heterostructures can be prepared by this strategy. Xia *et al.* [30] used the solution deposition method for *in situ* growth of MnO_2 nanosheets on the surface of g- C_3N_4 nanolayers. As shown in Fig. 5a, the g- C_3N_4 nanosheets were obtained after exfoliation, which possessed a large number of defects, such as dangling bonds, hydroxyl groups, and exposed lone pair electrons from N atoms. These defects promoted the adsorption of Mn^{2+} on the surface of g- C_3N_4 nanosheets. Then, Mn^{2+} would be gradually oxidized to generate MnO_2 na-

Table 1 *In situ* growth of 2D/2D heterostructures through wet chemical method

2D/2D heterojunction	Pre-synthesized 2D component/method	Growth method for the second 2D component	References
g-C ₃ N ₄ -GO/MoS ₂	g-C ₃ N ₄ -GO/Hydrothermal	Hydrothermal	[36]
TiO ₂ /GO	GO/Modified Hummer's method	Solvothermal	[37]
C ₃ N ₄ /Bi ₂₀ TiO ₃₂	C ₃ N ₄ /Ultrasonic exfoliation	Hydrothermal + Calcination	[26]
CdS/rGO	GO/Modified Hummer's method	Hydrothermal + Hydrazine hydrate reduction	[38]
GO/Mesoporous TiO ₂	GO/Modified Hummer's method	Hydrothermal + Calcination	[39]
MoS ₂ /TiO ₂	TiO ₂ /Hydrothermal	Hydrothermal	[40]
BiOI ₃ /BiOI	BiOI ₃ /hydrothermal	Chemical precipitation method	[41]
P-C ₃ N ₄ /ZnIn ₂ S ₄	P-C ₃ N ₄ /Frozen expansion and post-thermal exfoliation	Hydrothermal	[42]
TiO ₂ /Bi ₂ WO ₆	TiO ₂ /Hydrothermal	Hydrothermal	[43]
MnO ₂ /rGO	rGO/Modified Hummers' method + Calcination	Hydrothermal	[44]
BiOBr/La ₂ Ti ₂ O ₇	La ₂ Ti ₂ O ₇ /Hydrothermal	Chemical precipitation	[45]
BiOCl/La ₂ Ti ₂ O ₇	La ₂ Ti ₂ O ₇ /Hydrothermal	Chemical precipitation	[46]
BiOI/g-C ₃ N ₄	g-C ₃ N ₄ /Ultrasonic exfoliation	Chemical precipitation	[47]
MnO ₂ /g-C ₃ N ₄	g-C ₃ N ₄ /Thermal exfoliation	Redox reaction	[48]
MnO ₂ /g-C ₃ N ₄	g-C ₃ N ₄ /Nitric acid and hydrogen peroxide exfoliation	Redox reaction	[30]
Bi ₄ Ti ₃ O ₁₂ /I-BiOCl	Bi ₄ Ti ₃ O ₁₂ /Molten salt synthesis	Chemical transformation	[31]
TiO ₂ /SnS ₂	TiO ₂ /Hydrothermal	Hydrothermal	[49]
Bi ₂ WO ₆ /RGO/g-C ₃ N ₄	rGO/g-C ₃ N ₄ /heat-etchin + Hydrothermal	Hydrothermal	[50]
Ti ₃ C ₂ /Bi ₂ WO ₆	Ti ₃ C ₂ /Ultrasonic exfoliation	Hydrothermal	[28]
N-ZnO-g-C ₃ N ₄	g-C ₃ N ₄ /Ultrasonic exfoliation	Hydrothermal	[51]
N-ZnO-g-C ₃ N ₄	g-C ₃ N ₄ /Ultrasonic exfoliation	Hydrothermal	[52]
g-C ₃ N ₄ /ZnIn ₂ S ₄	g-C ₃ N ₄ /Thermal exfoliation	Surfactant-assisted solvothermal	[53]
SnNb ₂ O ₆ /CoFe-LDH	SnNb ₂ O ₆ /Hydrothermal	Hydrothermal	[54]
BiOI/BiVO ₄	BiOI/Hydrolysis	Anion-exchange	[55]
Cu/TiO ₂ @Ti ₃ C ₂ Tx	Ti ₃ C ₂ Tx/Etching	Hydrothermal	[56]
g-C ₃ N ₄ /NiAl-LDH	g-C ₃ N ₄ /Ultrasonic exfoliation	Hydrothermal	[57]
BiOCl/g-C ₃ N ₄	g-C ₃ N ₄ /Ultrasonic exfoliation	Hydrothermal	[58]
Zn ₃ In ₂ S ₆ /F-g-C ₃ N ₄	F-g-C ₃ N ₄ /Hydrothermal	Hydrothermal	[59]
CoMoS ₂ /rGO/C ₃ N ₄	g-C ₃ N ₄ /Thermal exfoliation rGO/Hummers' method and reduction	Solvothermal	[60]
MoS ₂ /CdS	CdS/Hydrothermal	Hydrothermal	[61]
g-C ₃ N ₄ /rGO/MoS ₂	g-C ₃ N ₄ -rGO/Pyrolysis	Hydrothermal	[62]
CuInS ₂ /SnS ₂	SnS ₂ /Hydrothermal	Hydrothermal	[63]
ZnCr-CLDH/g-C ₃ N ₄	g-C ₃ N ₄ /Thermal exfoliation	Chemical precipitation	[64]
g-C ₃ N ₄ /Bi ₁₂ O ₁₇ C ₁₂	g-C ₃ N ₄ /Thermal exfoliation	Chemical precipitation	[65]
MnIn ₂ S ₄ /g-C ₃ N ₄	g-C ₃ N ₄ /Frozen expansion and post-thermal exfoliation	Hydrothermal	[66]
CuInS ₄ /ZnIn ₂ S ₄	ZnIn ₂ S ₄ /Hydrothermal	Hydrothermal	[67]
CuInS ₄ /g-C ₃ N ₄	g-C ₃ N ₄ /Thermal exfoliation	Hydrothermal	[68]
C ₃ N ₄ /SnS ₂	g-C ₃ N ₄ /Ultrasonic exfoliation	Hydrothermal	[69]
MoS ₂ /SnNb ₂ O ₆	SnNb ₂ O ₆ /Hydrothermal	Hydrothermal	[70]
SnNb ₂ O ₆ /Bi ₂ WO ₆	SnNb ₂ O ₆ /Hydrothermal	Hydrothermal	[71]
ZnIn ₂ S ₄ /BiOCl	ZnIn ₂ S ₄ /Hydrothermal	Hydrothermal	[72]
P-La ₂ Ti ₂ O ₇ /Bi ₂ WO ₆	P-La ₂ Ti ₂ O ₇ /Hydrothermal + Calcination	Solvothermal	[73]
MoS ₂ /PbS	MoS ₂ /Liquid-phase exfoliation	Solvothermal	[74]
Ti ₃ C ₂ @TiO ₂ @MoS ₂	Ti ₃ C ₂ /HF Etching Ti ₃ C ₂ /TiO ₂ /Hydrothermal	Hydrothermal	[75]
BiOCl/K ⁺ Ca ₂ Nb ₃ O ₁₀ ⁻	K ⁺ Ca ₂ Nb ₃ O ₁₀ ⁻ /Solid-phase reaction	Hydrothermal	[76]
CdIn ₂ S ₄ /N-rGO	N-rGO/Ultrasonic treatment	Hydrothermal	[77]
SnS ₂ /TiO ₂	TiO ₂ /Hydrothermal	Hydrothermal	[78]
WS ₂ /TiO ₂	TiO ₂ /Hydrothermal	Hydrothermal	[79]
GO/Bi ₂ WO ₆	GO/Hummers' method and reduction	Hydrothermal	[80]
MoS ₂ /g-C ₃ N ₄	g-C ₃ N ₄ /Liquid exfoliation	Solvothermal	[81]
Black phosphorus (BP)/MoS ₂	BP/Ultrasonic exfoliation	Solvothermal	[82]
SnS ₂ /MoS ₂	SnS ₂ /Hydrothermal	Hydrothermal	[83]
Zn _x Cd _{1-x} In ₂ S ₄ /g-C ₃ N ₄	g-C ₃ N ₄ /Thermal exfoliation	Hydrothermal	[84]
CdS/g-C ₃ N ₄	g-C ₃ N ₄ /Liquid exfoliation	Hydrothermal	[85]

Figure 4 (a) Schematic illustration of the synthetic process for 2D/2D $\text{Ti}_3\text{C}_2/\text{Bi}_2\text{WO}_6$ heterojunction. DMSO: dimethyl sulfoxide. Reprinted with permission from Ref. [28]. Copyright 2018, Wiley-VCH. (b) Schematic illustration of the synthetic process for 2D/2D $\text{C}_3\text{N}_4/\text{Bi}_{20}\text{TiO}_{32}$ heterojunction. Reprinted with permission from Ref. [29]. Copyright 2015, Royal Society of Chemistry.

nosheets by addition of tetramethylammonium hydroxide (TMA-OH) and H_2O_2 in the suspension, and thus the 2D/2D g- $\text{C}_3\text{N}_4/\text{MnO}_2$ heterostructured photocatalyst was synthesized (Fig. 5b). In another study, Qian and coworkers [31] successfully constructed $\text{Bi}_4\text{Ti}_3\text{O}_{12}/\text{I-BiOCl}$ 2D/2D heterojunction systems *via* an *in situ* ion exchange approach at room temperature. As displayed in Fig. 5c, the pre-prepared $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ nanosheets served as the substrate. By mixing with HCl, the outside of the $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ nanosheets could be dissolved to Bi^{3+} and Ti^{4+} , and then an intermediate product BiO^+ was generated because of the hydrolysis of Bi^{3+} . Finally, the second phase I-BiOCl on the surface of the $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ nanosheets was gradually formed *via* the combination of BiO^+ , Cl^- , and I^- , and thus the $\text{Bi}_4\text{Ti}_3\text{O}_{12}/\text{I-BiOCl}$ 2D/2D heterojunction was synthesized (Fig. 5d).

Besides the wet-chemical method, CVD technique was also employed for *in-situ* growth of the second 2D component on the primarily synthesized 2D component. Typically, the pre-synthesized 2D substrate exposed to volatile precursors, which would react or decompose on the surface of substrate for growth of the second 2D component. Gong *et al.* [32] used the CVD method for growing WSe_2 on the edge and top surface of pre-synthesized MoSe_2 , resulting in the formation of $\text{WSe}_2/\text{MoSe}_2$ 2D/2D heterostructures. In another example, Han *et al.* [33] reported the *in-situ* fabrication of ultrathin SiC nanosheets on the 2D surface of reduced graphene oxide (rGO) through a facial CVD technique. The growth mechanism of SiC nanosheets was attributed to a vapor-solid reaction between Si atoms and solid carbon template.

Figure 5 (a, b) Schematic illustration of the synthetic process (a) and representative HRTEM image (b) of 2D/2D g-C₃N₄/MnO₂ heterojunction. Reprinted with permission from Ref. [30]. Copyright 2017, American Chemical Society. (c, d) Schematic illustration of the synthetic process (c) and representative TEM image (d) of 2D/2D Bi₄Ti₃O₁₂/I-BiOCl heterojunction. Reprinted with permission from Ref. [31]. Copyright 2017, Royal Society of Chemistry.

The 2D/2D heterostructures can also be obtained by a direct synthesis method without the need of pre-synthesized 2D substrate. For example, Zhang *et al.* [34] developed a modified nanoconfinement method for the direct synthesis of atomic-scale 2D/2D heterojunctions of boron nitride and monolayer graphene. In a typical procedure, the 2D/2D heterojunctions were directly synthesized by pyrolyzing the mixture of glucose (carbon source), boric acid (boron source), and urea (leaving reagent) at the elevated temperature of 1000°C under nitrogen atmosphere. More recently, Shi and coworkers [35] designed Cu₂S/Zn_{0.67}Cd_{0.33}S 2D/2D nanosheet heterojunctions through a one-step *in situ* topotactic hydrothermal transformation of CuZnCdAl layered double hydroxide (LDH) precursors.

Ex situ assembly of 2D/2D heterojunction photocatalysts

Compared with the aforementioned *in situ* growth method, each component in 2D/2D heterojunction is separately synthesized in this strategy. Then, these 2D pre-synthesized components are combined together through some assembly strategies, mainly including liquid-phase ultrasonic adsorption, solid phase grinding, electrostatic attraction, hydrothermal and calcination, which are summarized in Table 2 [86–125].

Liquid-phase ultrasonic adsorption is one of the simplest routes, which is often carried by a direct ultrasonic treatment of the mixed suspension of the 2D components. Ma *et al.* [86] constructed 2D CdS/MoS₂ heterojunction by ultrasonic adsorption of CdS nanosheets and

MoS₂ nanosheets. As shown in Fig. 6a, the CdS nanosheets and MoS₂ nanosheets were firstly synthesized by the solvothermal and hydrothermal method, respectively. Then, a certain amount of the CdS nanosheets as well as MoS₂ nanosheets were dispersed into water to form a mixed suspension. The CdS/MoS₂ composites were obtained after the ultrasonic treatment of the mixed suspension and followed vacuum-drying. In another study, Qiao's group [87] fabricated a metal-free phosphorene (FP)/graphitic carbon nitride (CNS) 2D/2D heterojunction through mechanically grinding the FP/CNS ethanol suspension. The TEM image (Fig. 6b) clearly revealed that the 2D CNS was attached on the surface of ultrathin FP based on weak vdW force. Solid phase grinding along with post-sintering process was also employed to assemble the 2D/2D heterojunction. Xu and coworkers [88] successfully synthesized a Bi₄NbO₈Cl/g-C₃N₄ 2D/2D heterojunction *via* the solid phase grinding and post-sintering process. As shown in Fig. 6c, Bi₄NbO₈Cl and g-C₃N₄ nanosheets were firstly prepared by a molten-salt and thermal polymerization method, respectively. Then, the Bi₄NbO₈Cl and g-C₃N₄ nanosheets mixed together by ball-milling as well as further calcination in air. Similarly, Liu *et al.* [89] also used this strategy to fabricate 2D/2D Bi₄Ti₃O₁₂/Ni(OH)₂ composites.

Although the coupling of 2D components *via* liquid-phase ultrasonic adsorption or solid phase grinding is a simple and direct route, the interface adhesion in such obtained 2D/2D systems is usually not strong, which may result in the breaking down of their structures in practical

Table 2 Summary of the *ex situ* methods to assembly of 2D/2D heterojunction

2D/2D heterojunction	Component A/Method	Component B/Method	Assembly methods	References
SnNb ₂ O ₆ /Graphene	SnNb ₂ O ₆ /(Hydrothermal + Calcination + Positively-charged functionalization)	Graphene/(Modified Hummers method + BPEI refluxing)	Electrostatic attraction	[95]
rGO/g-C ₃ N ₄	g-C ₃ N ₄ /(Thermal polymerization + Ultrasonic exfoliation + Proton-functionalized)	rGO/(Modified Hummers method + Ultrasonic exfoliation + NaBH ₄ reduction)	Electrostatic attraction	[96]
SnS ₂ /g-C ₃ N ₄	SnS ₂ /Hydrothermal	g-C ₃ N ₄ /(Thermal polymerization + Ultrasonic exfoliation)	Ultrasonic adsorption + Hydrothermal	[97]
C ₃ N ₄ /rGO	C ₃ N ₄ /(Thermal polymerization + Ultrasonic exfoliation + Proton-functionalized)	rGO/Modified Hummers method	Photo-assisted electrostatic attraction	[98]
Bi ₂ O ₂ CO ₃ /g-C ₃ N ₄	Bi ₂ O ₂ CO ₃ /Hydrothermal	g-C ₃ N ₄ /Thermal polymerization	Calcination	[99]
MoS ₂ /g-C ₃ N ₄	MoS ₂ /Hydrothermal + Ultrasonic exfoliation	g-C ₃ N ₄ /(Thermal polymerization + Ultrasonic exfoliation)	Impregnation and calcination method	[100]
GL-MoS ₂ /C ₃ N ₄	MoS ₂ /Hydrothermal	C ₃ N ₄ /Thermal polymerization	Hydrothermal	[101]
SnNb ₂ O ₆ /g-C ₃ N ₄	SnNb ₂ O ₆ /Hydrothermal	g-C ₃ N ₄ /(Thermal polymerization + HNO ₃ exfoliation)	Hydrothermal	[102]
C ₃ N ₄ /GO	C ₃ N ₄ /Thermal polymerization	GO/Modified Hummers method	Ultrasonic adsorption + Freeze drying	[103]
GO/g-C ₃ N ₄	GO/Modified Hummers method	g-C ₃ N ₄ /(Thermal polymerization + Ultrasonic exfoliation + Proton-functionalized)	Photo-assisted electrostatic attraction	[104]
Bi ₂ WO ₆ /TiO ₂	TiO ₂ /(Hydrothermal + Positively-charged functionalization)	Bi ₂ WO ₆ /(Hydrothermal + A-TNS functionalization)	Electrostatic attraction	[91]
g-C ₃ N ₄ /K ⁺ Ca ₂ Nb ₃ O ₁₀ ⁻	K ⁺ Ca ₂ Nb ₃ O ₁₀ ⁻ /TBA ⁺ OH ⁻ ultrasonic exfoliation	g-C ₃ N ₄ /Thermal polymerization	Hydrothermal	[93]
WO ₃ /K ⁺ Ca ₂ Nb ₃ O ₁₀ ⁻	WO ₃ /Hydrothermal	K ⁺ Ca ₂ Nb ₃ O ₁₀ ⁻ /TBAOH ultrasonic exfoliation	Hydrothermal	[94]
N-doped La ₂ Ti ₂ O ₇ /g-C ₃ N ₄	N-doped La ₂ Ti ₂ O ₇ /Hydrothermal	g-C ₃ N ₄ /(Thermal polymerization + Thermal exfoliation)	Ultrasonic adsorption	[105]
CdS/MoO ₂	CdS/Solvothermal	MoO ₂ /Hydrothermal	Ultrasonic adsorption	[86]
WO ₃ /SnNb ₂ O ₆	WO ₃ /Hydrothermal	SnNb ₂ O ₆ /Hydrothermal	Hydrothermal	[106]
BiOI/CeO ₂	CeO ₂ /Refluxed method	BiOI/Precipitation	Ultrasonic adsorption	[107]
BP/g-C ₃ N ₄	BP/NMP solvent exfoliation	g-C ₃ N ₄ /(Thermal polymerization + Thermal exfoliation)	Ultrasonic adsorption	[108]
BiOI ₃ /g-C ₃ N ₄	g-C ₃ N ₄ /(Thermal polymerization + Ultrasonic exfoliation)	BiOI ₃ (Hydrothermal + Positively-charged functionalization)	Electrostatic attraction	[109]
rGO/g-C ₃ N ₄	g-C ₃ N ₄ /(Thermal polymerization + Ultrasonic exfoliation + Proton-functionalized)	GO/Modified Hummers method	Electrostatic attraction	[110]
Porous-g-C ₃ N ₄ /Bi ₂ WO ₆	Porous-g-C ₃ N ₄ /Thermal polymerization	Bi ₂ WO ₆ /Hydrothermal	Ultrasonic adsorption	[111]
Ni ₂ P/ZnIn ₂ S ₄	ZnIn ₂ S ₄ /Hydrothermal + Ultrasonic exfoliation	Ni ₂ P/Hydrothermal + Calcination	Ultrasonic adsorption	[112]
CdS/WS ₂	CdS/Hydrothermal	WS ₂ /NMP ultrasonic exfoliation	Stirring adsorption	[113]
MoO ₂ /GL-C ₃ N ₄	MoO ₂ /Interfacial self-assembly and thermal reduction	g-C ₃ N ₄ /(Thermal polymerization + Thermal exfoliation)	Hydrothermal	[114]
Phosphorene/g-C ₃ N ₄	Phosphorene/Ultrasonic exfoliation	g-C ₃ N ₄ /(Thermal polymerization + Thermal exfoliation)	Mechanically ground	[87]
Fe ₂ O ₃ /g-C ₃ N ₄	Fe ₂ O ₃ /Hydrothermal	g-C ₃ N ₄ /(Thermal polymerization + Ultrasonic exfoliation + Proton-functionalized)	Electrostatic attraction	[92]
g-C ₃ N ₄ /MoS ₂	g-C ₃ N ₄ /(Thermal polymerization + Thermal exfoliation)	MoS ₂ /Ultrasonic exfoliation	Ultrasonic adsorption	[115]
WO ₃ /g-C ₃ N ₄	g-C ₃ N ₄ /(Thermal polymerization + Ultrasonic exfoliation + Positively-charged functionalization)	WO ₃ /BSA electrostatic-assisted ultrasonic exfoliation	Electrostatic attraction	[116]

(Continued)

2D/2D heterojunction	Component A/Method	Component B/Method	Assembly methods	References
ZnIn ₂ S ₄ /MoS ₂	ZnIn ₂ S ₄ /(Hydrothermal + Cryodesiccation)	MoS ₂ /(Hydrothermal + Cryodesiccation)	Electrostatic attraction	[117]
Ti ₃ C ₂ MXene/O-doped g-C ₃ N ₄	g-C ₃ N ₄ /(Thermal polymerization + Ultrasonic exfoliation + Calcination + Proton-functionalized)	Ti ₃ C ₂ /(HF etching + Ultrasonic exfoliation)	Electrostatic attraction	[118]
Ti ₃ C ₂ /g-C ₃ N ₄	Ti ₃ C ₂ /Pyroreaction + HF etching	g-C ₃ N ₄ /(Thermal polymerization + Thermal exfoliation + Calcination + Proton-functionalized)	Electrostatic attraction	[119]
BiVO ₄ /g-C ₃ N ₄	BiVO ₄ /Hydrothermal	g-C ₃ N ₄ /Thermal polymerization	Ultrasonic adsorption	[120]
WO ₃ /ZnIn ₂ S ₄	WO ₃ /(Hydrothermal + Calcination + Positively-charged functionalization)	ZnIn ₂ S ₄ /(Fe fluxing + Ultrasonic exfoliation)	Electrostatic attraction	[90]
ZnIn ₂ S ₄ /g-C ₃ N ₄	g-C ₃ N ₄ /(Thermal polymerization + Ultrasonic exfoliation + Calcination + Proton-functionalized)	ZnIn ₂ S ₄ /(Fe fluxing + Ultrasonic exfoliation)	Electrostatic attraction	[121]
ZnV ₂ O ₆ /g-C ₃ N ₄	ZnV ₂ O ₆ /(Hydrothermal + Calcination)	g-C ₃ N ₄ /(Thermal polymerization + Proton-functionalized)	Electrostatic attraction	[122]
SnS ₂ /g-C ₃ N ₄	g-C ₃ N ₄ /(Thermal polymerization + Thermal exfoliation)	SnS ₂ /Hydrothermal	Hydrothermal	[123]
BP/g-C ₃ N ₄	BP/Solvent exfoliation	g-C ₃ N ₄ /(Thermal polymerization + Thermal exfoliation)	Ultrasonic adsorption	[124]
Bi ₄ Ti ₃ O ₁₂ /Ni(OH) ₂	Bi ₄ Ti ₃ O ₁₂ /Molten salt method	Ni(OH) ₂ /Sedimentation	Solid phase grinding	[89]
Bi ₄ NbO ₈ Cl/g-C ₃ N ₄	Bi ₄ NbO ₈ Cl/Molten salt method	g-C ₃ N ₄ /Thermal polymerization	Solid phase grinding + Calcination	[88]
Bi ₂ WO ₆ /BiOI	Bi ₂ WO ₆ /Hydrothermal	BiOI/Sedimentation	Ultrasonic adsorption	[125]

Abbreviations: A-TNS: anatase TiO₂ nanosheet; BPEI: branched polyethylenimine; BSA: bovine serum albumin; GL: graphene-like; NMP: *N*-methyl-2-pyrrolidone.

Figure 6 (a) Schematic illustration of the synthetic process of 2D/2D CdS/MoS₂ heterojunction. DETA: diethylenetriamine. Reprinted with permission from Ref. [86]. Copyright 2017, Elsevier. (b) TEM image of metal-FP/graphitic CNS 2D/2D heterojunction. Reprinted with permission from Ref. [87]. Copyright 2018, Wiley-VCH. (c) Schematic illustration of the synthetic process of 2D/2D Bi₄NbO₈Cl/g-C₃N₄ heterojunction. Reprinted with permission from Ref. [88]. Copyright 2019, Elsevier.

applications. Thus, some approaches have been developed to consolidate their heterojunction interfaces. One of the most effective strategies is to respectively modify each 2D component with heterogeneous charges. Then, an electrostatic attraction occur between them when they mix together, which benefits the fabrication of the 2D/2D heterojunction with good interfacial contact. According to this route, a variety of advanced 2D/2D heterojunctions have been synthesized, which are summarized in Table 2. For example, Tan *et al.* [90] constructed 2D/2D $\text{WO}_3/\text{ZnIn}_2\text{S}_4$ nanocomposites *via* electrostatic attraction between modified WO_3 and ZnIn_2S_4 . As shown in Fig. 7a, WO_3 nanosheets were synthesized through a hydrothermal process and post-annealing treatment while the negatively charged ZnIn_2S_4 nanosheets were obtained by a refluxing and ultrasonic exfoliation process. The WO_3 nanosheets were further modified by adding 3-aminopropyltriethoxysilane (APTES) to obtain positively-charged WO_3 disperse solution, and then the negatively charged ZnIn_2S_4 nanosheets were added into the suspension to assemble 2D/2D $\text{WO}_3/\text{ZnIn}_2\text{S}_4$ heterostructure. TEM (Fig. 7b) and HRTEM (Fig. 7c) images revealed the intimate contact interfaces between WO_3 and ZnIn_2S_4 nanosheets. Yuan *et al.* [91] also employed the

electrostatic self-assembly method to design a 2D/2D heterojunction. The positively charged TiO_2 nanosheets and negatively charged Bi_2WO_6 were combined together to couple the 2D/2D $\text{Bi}_2\text{WO}_6/\text{TiO}_2$ heterojunction (Fig. 7d). In another study, Xu and coworkers [92] synthesized 2D/2D $\text{Fe}_2\text{O}_3/\text{g-C}_3\text{N}_4$ composites *via* the facile electrostatic self-assembly approach. As confirmed by the measured Zeta potential results, the strong negative charge on $\text{g-C}_3\text{N}_4$ could provide a significant driving force to attract positively charged Fe_2O_3 , resulting in spontaneous assembly of the two components into a 2D/2D heterostructure (Fig. 7e).

Hydrothermal co-assembly is also an effective method to couple the pre-synthesized 2D components. The formed 2D/2D heterostructure also possesses strong contact interface. Jiang *et al.* [93] coupled the perovskite oxide ultrathin nanosheets with $\text{g-C}_3\text{N}_4$ nanosheets by the hydrothermal coassembly method. As displayed in Fig. 8a, ultrathin $\text{K}^+\text{Ca}_2\text{Nb}_3\text{O}_{10}^-$ nanosheets were obtained by exfoliation of the $\text{KCa}_2\text{Nb}_3\text{O}_{10}$ bulk materials in tetrabutylammonium hydroxide (TBAOH) aqueous solution and the $\text{g-C}_3\text{N}_4$ nanosheets were prepared by a thermal polymerization strategy. Afterwards, a certain amount of the as-prepared $\text{g-C}_3\text{N}_4$ nanosheets and

Figure 7 (a–c) Schematic illustration of the synthetic process (a), TEM image (b) and HRTEM image (c) of the 2D/2D $\text{WO}_3/\text{ZnIn}_2\text{S}_4$ heterojunction. Reprinted with permission from Ref. [90]. Copyright 2019, Royal Society of Chemistry. (d) TEM image of the 2D/2D $\text{Bi}_2\text{WO}_6/\text{TiO}_2$ heterojunction. Reprinted with permission from Ref. [91]. Copyright 2017, Wiley-VCH. (e) TEM image of 2D/2D $\text{Fe}_2\text{O}_3/\text{g-C}_3\text{N}_4$ composites. Reprinted with permission from Ref. [92]. Copyright 2018, Wiley-VCH.

Figure 8 (a) Schematic illustration of the synthetic process of 2D/2D $\text{g-C}_3\text{N}_4/\text{K}^+\text{Ca}_2\text{Nb}_3\text{O}_{10}^-$ heterojunction. Reprinted with permission from Ref. [93]. Copyright 2017, Elsevier. (b, c) TEM and HRTEM images of 2D/2D $\text{WO}_3/\text{K}^+\text{Ca}_2\text{Nb}_3\text{O}_{10}^-$ heterojunction. Reprinted with permission from Ref. [94]. Copyright 2017, Royal Society of Chemistry.

$\text{K}^+\text{Ca}_2\text{Nb}_3\text{O}_{10}^-$ nanosheets were simultaneously added in deionized water to form a homogeneous suspension. The 2D/2D $\text{g-C}_3\text{N}_4/\text{K}^+\text{Ca}_2\text{Nb}_3\text{O}_{10}^-$ heterostructure (Fig. 8a) was fabricated by a facile one-step hydrothermal treatment of the mixture suspension. Similarly, Ma and coworkers [94] further coupled the ultrathin $\text{K}^+\text{Ca}_2\text{Nb}_3\text{O}_{10}^-$ nanosheets with WO_3 nanosheets *via* the hydrothermal coassembly route. The TEM image (Fig. 8b) and HRTEM (Fig. 8c) image confirmed that the WO_3 nanosheets and $\text{K}^+\text{Ca}_2\text{Nb}_3\text{O}_{10}^-$ nanosheets successfully combined each other by forming an intimate 2D/2D heterojunction interface.

APPLICATIONS OF 2D/2D HETEROJUNCTION PHOTOCATALYSTS

The 2D/2D heterojunction materials can display huge advantages toward photocatalytic reactions including the preferable dimensionality design, appropriate band structure, and surface properties. To date, the advanced 2D/2D heterojunction photocatalysts have been widely applied in diverse photocatalytic applications. In this section, the advancement of versatile photocatalytic applications of 2D/2D heterojunction photocatalysts in the

fields of hydrogen generation, environmental purification and CO_2 reduction will be discussed (Table 3).

Hydrogen generation

Hydrogen fuel has been considered as a clean and sustainable form of energy with the advantages including high energy density (142 MJ kg^{-1}), high stability and clean combustion product [126,127]. At present, the process of hydrogen generation mainly involves chemical decomposition of biomass [128], electrolysis of water [129], and photocatalytic decomposition of water [130–132]. Among these, hydrogen generation *via* photocatalytic water splitting is a promising eco-friendly route to address the energy crisis [133]. In this technology, it only needs the sustainable solar light as energy input, photocatalysts as medium, and water as reaction source, while there is no pollutive emission in the whole process. Thus, photocatalytic water splitting into hydrogen has been recognized as the “Holy Grail” of the renewable energy research. Up to now, various impressive photocatalyst materials have been developed for efficient and stable photocatalytic hydrogen production [134–139]. However, most of the employed photocatalysts are still

Table 3 Summary of the photocatalytic performances by using 2D/2D heterojunction photocatalysts

Photocatalytic hydrogen generation							
Photocatalysts	Amount of photocatalysts/Reaction solution volume	Sacrificial agent	Light source	Yields of product ($\mu\text{mol h}^{-1}\text{g}^{-1}$)	Apparent quantum yield (AQY)	References	
MoS ₂ /g-C ₃ N ₄	20 mg/100 mL	Lactic acid	300 W Xe ($\lambda > 420$ nm)	1030	2.1% at 420 nm	[203]	
TiO ₂ /MoS ₂	100 mg/100 mL	Methanol	300 W Xe	2145	6.4% at 360 nm	[40]	
CdS-NSS/rGO-WO ₃	-/30 mL	EtOH	300 W Xe ($\lambda > 420$ nm)	119.4	10.7% at 420 nm	[152]	
g-C ₃ N ₄ /N-La ₂ Ti ₂ O ₇	5 mg/5 mL	Methanol	500 W Xe	430	2.1% at 420 nm	[105]	
TiO ₂ /g-C ₃ N ₄	10 mg/30 mL	TEOA	300 W Xe	18,200	5.3% at 380 nm	[141]	
ZnO-MoS ₂ /rGO	5 mg/-	Na ₂ S/Na ₂ SO ₃	Natural sunlight irradiation	28,616	-	[204]	
CdS/MoS ₂	50 mg/80 mL	Na ₂ S/Na ₂ SO ₃	300 W Xe ($\lambda > 400$ nm)	8720	-	[86]	
Bi ₄ Ti ₃ O ₁₂ /I-BiOCl	50 mg/80 mL	Methanol	350 W Xe ($\lambda > 420$ nm)	91.7	-	[31]	
α -Fe ₂ O ₃ /g-C ₃ N ₄	10 mg/100 mL	TEOA	300 W Xe ($\lambda > 400$ nm)	>30,000	44.35% at 420 nm	[150]	
CPFA/g-C ₃ N ₄	100 mg/300 mL	Triethanol amine	300 W Xe ($\lambda > 400$ nm)	584.7	-	[205]	
ZnIn ₂ S ₄ /MoSe ₂	5 mg/10 mL	Lactic acid	300 W Xe ($\lambda > 400$ nm)	6454	-	[206]	
Nickel boron oxide/Graphene	40 mg/100 mL	TEOA	300 W Xe ($\lambda > 400$ nm)	~5000	-	[207]	
MoS ₂ /Cu-ZnIn ₂ S ₄	50 mg/250 mL	Ascorbic acid	300 W Xe ($\lambda > 420$ nm)	5463	13.6% at 420 nm	[160]	
MoS ₂ /rGO	40 mg/250 mL	TEOA and [ZnTMPyP] ⁴⁺	300 W Xe ($\lambda > 420$ nm)	2560	15.2% at 420 nm	[208]	
BP/g-C ₃ N ₄	1.5 mg/40 mL	Methanol	320 W Xe ($\lambda > 420$ nm)	427	-	[108]	
N-ZnO/g-C ₃ N ₄	5 mg/50 mL	Na ₂ S/Na ₂ SO ₃	320 W Xe	18,836	-	[52]	
g-C ₃ N ₄ /MoS ₂	50 mg/80 mL	Methanol	350 W Xe ($\lambda > 400$ nm)	191.2	-	[209]	
Ni ₂ P/ZnIn ₂ S ₄	50 mg/100 mL	Lactic acid	300 W Xe ($\lambda > 400$ nm)	2066	7.7% at 420 nm	[112]	
g-C ₃ N ₄ /ZnIn ₂ S ₄	50 mg/60 mL	TEOA	300 W Xe ($\lambda > 420$ nm)	2780	7.05% at 420 nm	[53]	
Cu _y /TiO ₂ @Ti ₃ C ₂ Tx	20 mg/150 mL	Methanol	300 W Xe	860	-	[56]	
Phosphorene/g-C ₃ N ₄	20 mg/100 mL	Lactic acid	300 W Xe ($\lambda > 400$ nm)	571	1.2% at 420 nm	[87]	
g-C ₃ N ₄ /MgFe	30 mg/100 mL	Tricthanolamine	300 W Xe ($\lambda > 420$ nm)	1260	6.9% at 420 nm	[210]	
g-C ₃ N ₄ /MoS ₂	3 mg/5 mL	Lactic acid	350 W Xe ($\lambda > 400$ nm)	660	5.67% at 400 nm	[115]	
g-C ₃ N ₄ /rGO	100 mg/100 mL	Tricthanolamine	300 W Xe ($\lambda > 420$ nm)	715	-	[211]	
CoP/g-C ₃ N ₄	50 mg/100 mL	Tricthanolamine	300 W Xe ($\lambda > 400$ nm)	~750	4.3% at 420 nm	[212]	
Fe ₂ O ₃ /g-C ₃ N ₄	50 mg/80 mL	TEOA	350 W Xe ($\lambda > 420$ nm)	398.0	-	[92]	
CoMoS ₂ /rGO/C ₃ N ₄	100 mg/-	TEOA	300 W Xe ($\lambda > 400$ nm)	684	-	[60]	
CdS-MoS ₂ /rGO-E	20 mg/80 mL	Lactic acid	300 W Xe ($\lambda > 420$ nm)	36,700	30.5% at 420 nm	[213]	
MoS ₂ /CdS	50 mg/250 mL	Na ₂ S/Na ₂ SO ₃	300 W Xe ($\lambda > 420$ nm)	26,320	46.65% at 450 nm	[61]	
g-C ₃ N ₄ /Graphene/MoS ₂	50 mg/250 mL	TEOA	300 W Xe ($\lambda > 420$ nm)	317	3.4% at 420 nm	[62]	
CdS/WS ₂	3 mg/5 mL	Lactic acid	350 W Xe	14,100	70% at 460 nm	[113]	
O-g-C ₃ N ₄ /TiO ₂	50 mg/50 mL	TEOA	300 W Xe ($\lambda > 400$ nm)	587.1	-	[142]	
Phosphorus/Bismuth vanadate	5 mg/8 mL	-	320 W Xe ($\lambda > 420$ nm)	160	0.89% at 420 nm	[214]	
CdS/WS ₂ /g-C ₃ N ₄	10 mg/20 mL	TEOA	300 W Xe ($\lambda > 420$ nm)	1174.5	-	[215]	
WO ₃ /g-C ₃ N ₄	50 mg/80 mL	Lactic acid	350 W Xe	982	-	[116]	
CuInS ₂ /ZnIn ₂ S ₄	50 mg/100 mL	Na ₂ S/Na ₂ SO ₃	300 W Xe ($\lambda > 420$ nm)	3430.2	12.4% at 420 nm	[67]	
Phosphorus/Monolayer Bi ₂ WO ₆	20 mg/100 mL	TEOA	300 W Xe	21042	-	[153]	
La ₂ Ti ₂ O ₇ /In ₂ S ₃	60 mg/100 mL	Na ₂ S/Na ₂ SO ₃	300 W Xe ($\lambda > 400$ nm)	158.89	-	[144]	
ZnIn ₂ S ₄ /MoS ₂	-/40 mL	Lactic acid	300 W Xe ($\lambda > 400$ nm)	4974	-	[117]	
MoS ₂ /SnNb ₂ O ₆	50 mg/50 mL	Methanol	300 W Xe ($\lambda > 420$ nm)	258	-	[70]	
C ₃ N ₄ /MoS ₂	50 mg/100 mL	Methyl alcohol	300 W Xe	385.04	-	[216]	
Ti ₃ C ₂ MXene/MoS ₂	10 mg/-	TEOA	300 W Xe (AM 1.5)	6425.297	4.61% at 420 nm	[75]	
g-C ₃ N ₄ /UMOFNs	15 mg/30 mL	Actic acid	500 W Xe	1909.02	2.34% at 405 nm	[217]	

(Continued)

Photocatalytic hydrogen generation

Photocatalysts	Amount of photocatalysts/Reaction solution volume	Sacrificial agent	Light source	Yields of product ($\mu\text{mol h}^{-1} \text{g}^{-1}$)	Apparent quantum yield (AQY)	References
Ti ₃ C ₂ MXene/O-doped g-C ₃ N ₄	10 mg/80 mL	TEOA	300 W Xe	25,124	6.53% at 420 nm	[118]
Pt/g-C ₃ N ₄ /WO ₃	50 mg/50 mL	TEOA	300 W Xe ($\lambda > 420$ nm)	862	17.5% at 400 nm	[151]
Cu ₂ S/Zn _{0.67} Cd _{0.33} S	30 mg/-	Na ₂ S/Na ₂ SO ₃	300 W Xe ($\lambda > 420$ nm)	152,700	18.15% at 420 nm	[35]
Ti ₃ C ₂ /g-C ₃ N ₄	30 mg/40 mL	TEOA	200 W Hg	72.3	-	[119]
SnS ₂ /TiO ₂	20 mg/40 mL	Methanol	300 W Xe	652.4	-	[78]
WO ₃ /ZnIn ₂ S ₄	20 mg/100 mL	Na ₂ S/Na ₂ SO ₃	300 W Xe ($\lambda > 420$ nm)	2202.9	-	[80]
Ba ₅ Nb ₄ O ₁₅ /g-C ₃ N ₄	50 mg/100 mL	Oxalic acid	420 nm LEDs	26,700	6.1% at 420 nm	[143]
ZnIn ₂ S ₄ /g-C ₃ N ₄	10 mg/120 mL	TEOA	300 W Xe ($\lambda > 400$ nm)	8601.16	0.92% at 400 nm	[121]
MoS ₂ /g-C ₃ N ₄	50 mg/250 mL	TEOA	300 W Xe ($\lambda > 420$ nm)	1155	6.8% at 420 nm	[81]
BP/MoS ₂	10 mg/250 mL	Na ₂ S/Na ₂ SO ₃	300 W Xe ($\lambda > 420$ nm)	1286	1.2% at 420 nm	[82]
BP/g-C ₃ N ₄	10 mg/100 mL	TEOA	300 W Xe ($\lambda > 420$ nm)	384.17	-	[218]
BP/g-C ₃ N ₄	20 mg/100 mL	BPA	300 W Xe ($\lambda > 400$ nm)	259.04	-	[124]
Zn _x Cd _{1-x} In ₂ S ₄ /g-C ₃ N ₄	50 mg/50 mL	TEOA	300 W Xe ($\lambda > 420$ nm)	170.3	8.5% at 420 nm	[84]
CdS/CoP	20 mg/50 mL	Ethanol	300 W Xe	56,300	-	[219]
g-C ₃ N ₄ /Co@NC	10 mg/100 mL	TEOA	300 W Xe ($\lambda > 400$ nm)	1567	10.82 % at 400 nm	[220]
Co ₃ (PO ₄) ₂ /g-C ₃ N ₄	50 mg/100 mL	-	300 W Xe ($\lambda > 400$ nm)	375.6	1.32% at 420 nm	[221]
FeSe ₂ /g-C ₃ N ₄	30 mg/-	Na ₂ S/Na ₂ SO ₃	300 W Xe	1655.6	-	[222]
CdS/Cu ₇ S ₄	5 mg/80 mL	Na ₂ S/Na ₂ SO ₃	300 W Xe	278,000	14.7 % at 420 nm	[140]

Photocatalytic CO₂ reduction

Photocatalysts	Amount of photocatalysts	Light source	Main product	Yields of product ($\mu\text{mol h}^{-1} \text{g}^{-1}$)	References
rGO/g-C ₃ N ₄	100 mg	15 W energy-saving daylight	CH ₄	13.93	[96]
BiOI/g-C ₃ N ₄	100 mg	300 W Xe ($\lambda > 400$ nm)	CO	3.446	[47]
MnO ₂ /g-C ₃ N ₄	50 mg	300 W Xe	CO	2.04	[48]
ZnV ₂ O ₆ /rGO	100 mg	35 W HID Xe	CH ₃ OH	515.397	[223]
Ti ₃ C ₂ /Bi ₂ WO ₆	100 mg	simulated solar irradiation	CH ₄	1.78	[28]
			CH ₃ OH	0.44	
SiC/rGO	30 mg	300 W Xe	CH ₄	14.5425	[33]
α -Fe ₂ O ₃ /g-C ₃ N ₄	25 mg	300 W Xe	CO	6.85	[224]
Bi ₂ WO ₆ /rGO/g-C ₃ N ₄	50 mg	300 W Xe ($\lambda > 420$ nm)	CH ₄	2.51	[50]
			CO	15.96	
g-C ₃ N ₄ /NiAl-LDH	50 mg	300 W Xe ($\lambda > 420$ nm)	CO	8.2	[57]
ZnV ₂ O ₆ /g-C ₃ N ₄	100 mg	35 W HID Xe	CH ₃ OH	776	[122]
Bi ₂ WO ₆ /BiOI	-	500 W Xe ($\lambda > 400$ nm)	CH ₄	2.29	[125]
Bi ₄ NbO ₈ Cl/g-C ₃ N ₄	50 mg	300 W Xe	CO	2.26	[88]
MOF/rGO	40 mg	100 W LED lamp	CO	3.8 $\times 10^4$	[225]

Removal of pollutions

Photocatalysts	Amount of photocatalysts/Reaction solution volume	Target	Light source	Reaction time/De-gradation efficiency	Rate constant k (min^{-1})	References
g-C ₃ N ₄ /rGO	8.0 mg/5 mL	RhB	1000 W Xe ($\lambda > 400$ nm)	75 min/100%	0.063	[198]
		4-Nitrophenol		150 min/52%	-	
TiO ₂ /Graphene	40 mg/60 mL	RhB	500 W Hg	60 min/95%	0.046	[37]
		2,4-DCP		60 min/95%		
α -Fe ₂ O ₃ /Graphene	30 mg/-	RhB	350 W Xe	20 min/98%	0.19489	[226]
SnNb ₂ O ₆ /Graphene	30 mg/40 mL	RhB	300 W Xe ($\lambda > 420$ nm)	60 min/98%	0.0616	[95]
rGO/GdS	10 mg/30 mL	MB	$\lambda > 420$ nm	60 min/98%	0.068	[38]

(Continued)

Removal of pollutions

Photocatalysts	Amount of photocatalysts/Reaction solution volume	Target	Light source	Reaction time/De-gradation efficiency	Rate constant k (min^{-1})	References
$\text{C}_3\text{N}_4/\text{Bi}_{20}\text{TiO}_{32}$	100 mg/100 mL	RhB	300 W Xe ($\lambda > 420$ nm)	20 min/98%	-	[29]
Graphene/ TiO_2	10 mg/30 mL	MB	300 W Xe ($\lambda > 420$ nm)	3 h/~80%	0.00463	[39]
$\text{BiOI/O}_3/\text{BiOI}$	-	NO	$\lambda > 420$ nm	30 min/41.3%	-	[41]
$\text{SnS}_2/\text{g-C}_3\text{N}_4$	10 mg/100 mL	RhB	300 W Xe ($\lambda > 400$ nm)	20 min/99.8%	0.2	[97]
$\text{BiOBr}/\text{La}_2\text{Ti}_2\text{O}_7$	40 mg/100 mL	RhB	300 W Xe	-	0.092	[45]
$\text{BiOCl}/\text{La}_2\text{Ti}_2\text{O}_7$	40 mg/100 mL	RhB	300 W Hg	-	0.19	[46]
CNX-NSS/rGO	5 mg/10 mL	MB	300 W Xe	40 min/88%	-	[98]
$\text{P-C}_3\text{N}_4/\text{ZnIn}_2\text{S}_4$	30 mg/30 mL	4-Nitroaniline	300 W Xe ($\lambda > 400$ nm)	90 min/99.4%	-	[42]
$\text{Bi}_2\text{O}_2\text{CO}_3/\text{g-C}_3\text{N}_4$	50 mg/50 mL	RhB	500 W Xe ($\lambda > 420$ nm)	5 h/74%	-	[99]
$\text{MoS}_2/\text{g-C}_3\text{N}_4$	40 mg/50 mL	RhB	300 W Xe ($\lambda > 420$ nm)	20 min/96%	0.152	[100]
$\text{g-C}_3\text{N}_4/\text{Bi}_4\text{O}_5\text{I}_2$	500 mg/50 mL	RhB	$\lambda > 420$ nm	40 min/99%	0.06	[227]
$\text{C}_3\text{N}_4/\text{Graphene}$	20 mg/-	MO	300 W Xe ($\lambda > 420$ nm)	5 h/73%	-	[103]
$\text{MoS}_2/\text{g-C}_3\text{N}_4$	25 mg/50 mL	MO	300 W Xe ($\lambda > 400$ nm)	3 h/74.4%	0.461	[101]
$\text{SnNb}_2\text{O}_6/\text{g-C}_3\text{N}_4$	20 mg/80 mL	MB	500 W W ($\lambda > 420$ nm)	60 min/55%	-	[102]
$\text{g-C}_3\text{N}_4/\text{N-La}_2\text{Ti}_2\text{O}_7$	10 mg/10 mL	MO	300 W Xe ($\lambda > 400$ nm)	3 h/44%	-	[105]
$\text{g-C}_3\text{N}_4/\text{MgIn}_2\text{S}_4$	15 mg/30 mL	MO	300 W Xe ($\lambda > 400$ nm)	70%	-	[228]
$\text{g-C}_3\text{N}_4/\text{TiO}_2$	10 mg/30 mL	MO	300 W Xe	15 min/98%	0.189	[141]
$\text{g-C}_3\text{N}_4/\text{Bi}_4\text{O}_3\text{Br}_2$	10 mg/100 mL	RhB	300 W Xe ($\lambda > 400$ nm)	75 min/91%	-	[229]
$\text{K}^+\text{Ca}_2\text{Nb}_3\text{O}_{10}^-/\text{g-C}_3\text{N}_4$	40 mg/40 mL	TC	500 W W ($\lambda > 420$ nm)	90 min/80%	0.0137	[93]
$\text{WO}_3/\text{K}^+\text{Ca}_2\text{Nb}_3\text{O}_{10}^-$	40 mg/40 mL	TC	250 W Xe	120 min/85.8%	0.0151	[94]
$\text{WO}_3/\text{SnNb}_2\text{O}_6$	40 mg/40 mL	RhB	500 W W	180 min/93.4%	0.015	[106]
$\text{Bi}_4\text{Ti}_3\text{O}_{12}/\text{I-BiOCl}$	50 mg/80 mL	MB	350 W Xe ($\lambda > 420$ nm)	180 min/90%	0.013	[31]
$\text{CeO}_2/\text{BiOCl}$	20 mg/20 mL	RhB	direct sunlight	60 min/89%	-	[107]
$\text{g-C}_3\text{N}_4/\text{Bi}_2\text{WO}_6$	10 mg/50 mL	Ibuprofen	300 W Xe ($\lambda > 420$ nm)	60 min/96.1%	0.052	[196]
$\text{g-C}_3\text{N}_4/\text{MnO}_2$	50 mg/50 mL	Phenol	300 W Xe	180 min/73.6%	0.033	[30]
$\text{Bi}_2\text{S}_3\text{-BiOCl}$	10 mg/50 mL	X-3B	300 W Xe ($\lambda > 400$ nm)	30 min/74.6%	0.096	[201]
$\text{Bi}_2\text{WO}_6/\text{TiO}_2$	10 mg/40 mL	4-Nitroaniline	300 W Xe	16 min/100%	-	[91]
$\text{TiO}_2/\text{SnS}_2$	20 mg/100 mL	MB	250 W Hg	60 min/44%	-	[49]
$\text{ZnO}/\text{V}_2\text{O}_5$	10 mg/25 mL	MB	300 W Xe ($\lambda > 400$ nm)	400 min/90%	0.0052	[230]
$\text{C}_3\text{N}_4\text{-CdS}$	500 mg/1000 mL	MO	300 W Xe	210 min/97%	-	[231]
$\text{Bi}_3\text{O}_4\text{Cl}/\text{g-C}_3\text{N}_4$	50 mg/100 mL	TC	250 W Xe ($\lambda > 420$ nm)	60 min/76%	0.0205	[197]
$\text{BiOI/O}_3/\text{g-C}_3\text{N}_4$	-/50 mL	2,4,6-Trichlorophenol	500 W Xe	2.5 h/92%	0.016	[109]
$\text{Bi}_2\text{WO}_6/\text{g-C}_3\text{N}_4$	50 mg/100 mL	RhB	500 W W ($\lambda > 420$ nm)	-	0.043	[111]
$\text{MoO}_2/\text{g-C}_3\text{N}_4$	50 mg/50 mL	RhB	300 W Xe ($\lambda > 420$ nm)	120 min/97.5%	-	[114]
$\text{N-ZnO}/\text{g-C}_3\text{N}_4$	20 mg/50 mL	RhB	visible light irradiation	180 min/97 %	0.0089	[51]
$\text{KTiNbO}_5/\text{g-C}_3\text{N}_4$	100 mg/-	RhB	300 W Xe ($\lambda > 420$ nm)	80 min/89.9 %	-	[232]
$\text{ZnO-ZnCr}_2\text{O}_4/\text{g-C}_3\text{N}_4\text{-C(N)}$	50 mg/80 mL	Congo red	500 W Xe ($\lambda > 400$ nm)	60 min/70 %	0.0387	[64]
$\text{SnNb}_2\text{O}_6/\text{CoFe-LDH}$	50 mg/50 mL	MO	500 W Xe	60 min/83.3%	-	[54]
$\text{BiOI}/\text{BiVO}_4$	30 mg/50 mL	RhB	Sunlamp ($\lambda > 400$ nm)	75 min/97%	0.0467	[55]
$\text{g-C}_3\text{N}_4/\text{Bi}_{12}\text{O}_{17}\text{Cl}_2$	30 mg/50 mL	RhB	300 W Xe ($\lambda > 400$ nm)	60 min/90%	0.353	[65]
$\text{BiOCl}/\text{g-C}_3\text{N}_4$	50 mg/100 mL	4-Chlorophenol	300 W Xe ($\lambda > 420$ nm)	120 min/95%	0.025	[58]
WC/WO_3	20 mg/20 mL	RhB	500 W Xe ($\lambda > 400$ nm)	-/89%	-	[202]
$\text{g-C}_3\text{N}_4/\text{rGO}$	-/20 mL	MO	300 W Xe ($\lambda > 400$ nm)	180 min/97%	-	[199]
$\text{Zn}_3\text{In}_2\text{S}_6/\text{F-C}_3\text{N}_4$	40 mg/100 mL	MO	300 W Xe ($\lambda > 420$ nm)	60 min/99%	0.07329	[59]
$\text{GO}/\text{g-C}_3\text{N}_4$	20 mg/50 mL	RhB	500 W Xe	-	0.0514	[110]

(Continued)

Removal of pollutions

Photocatalysts	Amount of photocatalysts/Reaction solution volume	Target	Light source	Reaction time/De-gradation efficiency	Rate constant k (min^{-1})	References
MoSe ₂ /Bi ₂ WO ₆	100 mg/-	Toluene	300 W Xe ($\lambda > 420$ nm)	180 min/80%	-	[233]
FeOCl/GO	50 mg/100 mL	RhB	sunlight	10 min/100%	0.32	[234]
SnS ₂ /CuInS ₂	30 mg/100 mL	MO	300 W Xe ($\lambda > 400$ nm)	60 min/99%	-	[63]
BP/g-C ₃ N ₄	20 mg/80 mL	RhB	300 W Xe ($\lambda > 420$ nm)	30 min/97%	0.288	[200]
Cu ₂ WS ₄ /g-C ₃ N ₄	50 mg/100 mL	Cr(VI)	300 W Xe ($\lambda > 420$ nm)	100 min/98.3%	0.04076	[193]
		TC		120 min/68.1%	-	
Ag-WO ₃ /g-C ₃ N ₄	100 mg/300 mL	RhB	500 W Xe ($\lambda > 420$ nm)	40 min/96.2%	0.053	[235]
MnIn ₂ S ₄ /g-C ₃ N ₄	30 mg/30 mL	TC	300 W Xe ($\lambda > 400$ nm)	120 min/100%	-	[66]
CuInS ₂ /g-C ₃ N ₄	50 mg/100 mL	TC	300 W Xe ($\lambda > 420$ nm)	60 min/83.7%	0.02583	[68]
C ₃ N ₄ /SnS ₂	30 mg/30 mL	MB	LED light ($\lambda = 410$ nm)	30 min/98.7%	0.08258	[69]
SnNb ₂ O ₆ /Bi ₂ WO ₆	50 mg/-	Quinolone anti-biotic	300 W Xe ($\lambda > 420$ nm)	60 min/~90%	0.0406	[71]
		Norfloracin (NOR)				
CQDs-ZnIn ₂ S ₄ /BiOCl	50 mg/100 mL	Antibiotics	300 W Xe ($\lambda > 420$ nm)	120 min/83.7%	0.014	[72]
CoAl-LDH/g-C ₃ N ₄ /rGO	50 mg/200 mL	Congo red	300 W halogen	30 min/99%	-	[236]
		TC		60 min/99%	-	
Carbon dots-BiVO ₄ /Bi ₃ TaO ₇	10 mg/30 mL	TC	500 W Xe ($\lambda > 420$ nm)	120 min/85.3%	-	[237]
MoS ₂ /PbS	25 mg/250 mL	MB	300 W Xe ($\lambda > 420$ nm)	48 min/90%	0.0431	[74]
Bi@Bi ₅ O ₇ /IrGO	30 mg/-	Levofloxacin	300 W Xe ($\lambda > 420$ nm)	60 min/87.7%	0.0322	[238]
BiOCl/K ⁺ Ca ₂ Nb ₃ O ₁₀ ⁻	35 mg/35 mL	TC	250 W Xe	150 min/94.5%	0.01568	[76]
CdIn ₂ S ₄ /N-rGO	50 mg/100 mL	2,4-DCP	$\lambda > 420$ nm	6 h/70%	0.0044	[77]
SnS ₂ /g-C ₃ N ₄	10 mg/50 mL	RhB	300 W Xe ($\lambda > 400$ nm)	60 min/94.8%	0.0302	[123]
BiVO ₄ /g-C ₃ N ₄	10 mg/20 mL	RhB	300 W Xe ($\lambda > 420$ nm)	60 min/100%	0.0410	[120]
Wg-C ₃ N ₄ /g-C ₃ N ₄	350 mg/125 mL	AV-7	fluorescent	30 min/96%	0.0809	[239]
TiO ₂ /WS ₂	20 mg/100 mL	RhB	300 W Xe ($\lambda > 420$ nm)	90 min/100%	-	[79]
rGO-BWO	20 mg/100 mL	TC	-	60 min/85.0%	0.030	[80]
SnS ₂ /MoS ₂	25 mg/100 mL	MB	250 W Hg	60 min/100%	0.00937	[83]
Ni(OH) ₂ /Bi ₄ Ti ₃ O ₁₂	20 mg/100 mL	Levofloxacin	300 W Xe	80 min/62%	0.01152	[89]
CdS/g-C ₃ N ₄	50 mg/50 mL	RhB	500 W Xe ($\lambda > 420$ nm)	120 min/96.5 %	0.02414	[85]
In ₂ S ₃ /Bi ₂ O ₂ CO ₃	30 mg/30 mL	RhB	400 W Xe	60 min/91 %	0.035	[156]

Abbreviations: AV-7: acid violet-7; 2,4-DCP: 2,4-dichlorophenol; LED: light-emitted diode; MB: methylene blue; MO: methyl orange; TEOA: triethanolamine; UMOFNS: ultrathinning metal-organic frameworks

subjected to low photocatalytic efficiency, which is far from enough to meet the requirements of practical applications. Generally, there are several shortcomings responsible for the low photocatalytic efficiency of the current photocatalysts, such as rapid recombination of photogenerated charge carriers, poor light harvesting and inactive charge transport kinetics [140].

Recent studies regarding 2D/2D heterojunction materials found that these composite photocatalysts with properly engineered energy band matching and preferable dimensionality design are quite promising to overcome the above-mentioned shortcomings in conventional photocatalysts, thus reaching the excellent hydrogen

evolution efficiency [140]. Many advanced 2D/2D heterojunction photocatalysts have been explored to split water for hydrogen generation as summarized in Table 3. Typically, the advanced 2D/2D composite semiconductors can be classified into five types, including Type-I, Type-II, Z-scheme, S-scheme and cocatalyst/photocatalyst (Fig. 9).

In a typical 2D/2D Type-I configuration (Fig. 9a), the valence band (VB) and conduction band (CB) of one 2D semiconductor (marked as B) are located within the band gap of another 2D semiconductor (marked as A). Thus, semiconductor A is often mainly responsible for harvesting light to produce electrons and holes, while semi-

Figure 9 Schematic diagrams of the five types of heterojunctions.

conductor B acts as electron as well as hole acceptors to promote the charge transfer. In 2017, Zhu *et al.* [108] developed an efficient metal-free 2D/2D heterojunction photocatalyst with Type-I configuration for photocatalytic H_2 evolution. The 2D/2D nanohybrid was composed of BP nanoflakes and $\text{g-C}_3\text{N}_4$ (CN) nanosheets. In this photocatalyst system, the CN was mainly excited to generate electrons and holes under visible light irradiation, while BP acted as an electron and hole acceptor from adjacent CN to inhibit the recombination of the photogenerated charges in CN (Fig. 10a). As a result, the 2D/2D CN/BP photocatalytic system with an optimal ratio of BP:CN at 1:4 presented a higher H_2 evolution rate of $427 \mu\text{mol g}^{-1} \text{h}^{-1}$. Another interesting 2D/2D metal-free photocatalyst with Type-I configuration was reported by Qiao's group [87]. They prepared a novel 2D/2D phosphorene/ $\text{g-C}_3\text{N}_4$ vdW heterojunction, which exhibited an enhanced visible-light photocatalytic H_2 production activity of $571 \mu\text{mol g}^{-1} \text{h}^{-1}$. 2D chalcogenide material can also be combined with $\text{g-C}_3\text{N}_4$ to construct 2D/2D Type-I heterojunction for excellent photocatalytic H_2 evolution. Lin *et al.* [53] reported 2D/2D $\text{g-C}_3\text{N}_4$ nanosheet/ ZnIn_2S_4 nanoleaf for photocatalytic H_2 generation. According to the Type-I charge transfer mechanism (Fig. 10b), the photoinduced CB electrons and VB holes on the $\text{g-C}_3\text{N}_4$ nanosheets could readily transfer to the CB and VB of ZnIn_2S_4 , respectively, contributing to fairly high photoinduced charge separation and migration efficiency. Furthermore, the unusual 2D/2D heterojunction

geometry endowed the heterojunction system with high-speed charge transfer nanochannels, ultimately resulting in a remarkable visible-light-driven H_2 evolution rate of $2780 \mu\text{mol g}^{-1} \text{h}^{-1}$. More recently, Yang *et al.* [121] made further progress by using 2D/2D $\text{g-C}_3\text{N}_4/\text{ZnIn}_2\text{S}_4$ Type-I heterojunction for photocatalytic H_2 evolution. In this study, they fabricated 2D/2D ultra-thin ZnIn_2S_4 /protonated $\text{g-C}_3\text{N}_4$ nanocomposites, which showed excellent photocatalytic H_2 production rate of $8601.16 \mu\text{mol g}^{-1} \text{h}^{-1}$ under visible light irradiation.

Type-II composite structures consist of two semiconductors with a staggered energy band alignment (Fig. 9b). During the photocatalytic process, the photoinduced electrons in the CB of semiconductor A transfer to the CB of semiconductor B, while the holes in the VB of A are transferred to the VB of B, which is derived by the potential between them. Compared with conventional Type-II heterojunctions, 2D Type-II heterojunctions own unique features in photocatalytic applications. The face-to-face manner not only offers the most charge transportation pathways but also ensures a very short migration distance for photogenerated charges, leading to a boosted photocatalytic performance. Gu *et al.* [141] reported a face-to-face interfacial assembly of $\text{TiO}_2/\text{g-C}_3\text{N}_4$ Type-II hybrid for photocatalytic hydrogen evolution. Under UV-Vis light irradiation, both $\text{g-C}_3\text{N}_4$ and TiO_2 can be excited by photon energy higher than their band-gaps. Then, the photoinduced electrons of the CB in $\text{g-C}_3\text{N}_4$ would drill into the CB of TiO_2 , while the photo-

Figure 10 (a) Schematic diagram for the photocatalytic H₂ evolution using BP/CN photocatalyst. Reprinted with permission from Ref. [108]. Copyright 2017, American Chemical Society. (b) Schematic diagram for the photocatalytic H₂ evolution using 2D/2D g-C₃N₄/ZnIn₂S₄ photocatalyst. NHE: normal hydrogen electrode. Reprinted with permission from Ref. [53]. Copyright 2018, Elsevier. (c) Schematic diagram for the photocatalytic H₂ evolution using La₂Ti₂O₇/In₂S₃ photocatalyst. Reprinted with permission from Ref. [144]. Copyright 2019, Elsevier.

induced holes of the VB in TiO₂ will transfer to the VB of g-C₃N₄. Due to the unique, ultrathin 2D face-to-face contact features, the photoinduced charge carriers would travel a very short distance between g-C₃N₄ and TiO₂, resulting in a high photocatalytic hydrogen evolution rate of 18,200 μmol g⁻¹ h⁻¹. Later, Zhong and coworkers [142] prepared a 2D/2D O-g-C₃N₄/TiO₂ composite with Type-II configuration for visible-light-driven photocatalytic hydrogen evolution. Some perovskite-type 2D semiconductors with a hexagonal crystal structure can also be coupled with 2D g-C₃N₄ to form a 2D/2D Type-II photocatalyst for photocatalytic hydrogen evolution. Cai *et al.* [105] reported the hybridization of 7 nm thick N-doped La₂Ti₂O₇ (NLTO) nanosheets with 2 nm thick g-C₃N₄ nanosheets to construct a g-C₃N₄/NLTO composite with a Type-II band alignment, in which the g-C₃N₄ and NLTO respectively act as hole receptor and electron

conductor. As a result, the optimal g-C₃N₄/NLTO photocatalyst exhibited a H₂ evolution rate of 430 μmol g⁻¹ h⁻¹, which was 10 and 2 times higher than those of LTO and NLTO, respectively. More recently, Wang *et al.* [143] employed another layered perovskite semiconductor (Ba₅Nb₄O₁₅) to combine with g-C₃N₄ to form a 2D/2D Type-II heterojunction towards enhanced photocatalytic activity. The Ba₅Nb₄O₁₅/g-C₃N₄ photocatalyst exhibited a higher hydrogen evolution rate of 2,670 μmol g⁻¹ h⁻¹, which was 2.35 times that of g-C₃N₄. Layered perovskite semiconductors have also been reported to couple with other 2D materials to form 2D/2D heterojunctions with Type-II configuration. For instance, Qian *et al.* [31] designed Bi₄Ti₃O₁₂/I-BiOCl 2D/2D heterojunction systems for photocatalytic hydrogen production. In another study, Hua and coworkers [144] fabricated a Type-II La₂Ti₂O₇/In₂S₃ heterojunction. As shown in Fig. 10c, the La₂Ti₂O₇/In₂S₃ nanosheet heterojunction possessed intimate face-to-face contact feature between them, which guarantees facile electron migrations from In₂S₃ to La₂Ti₂O₇. Consequently, the La₂Ti₂O₇/In₂S₃ nanosheet heterojunctions demonstrated a much improved photocatalytic hydrogen production compared with pristine In₂S₃ as well as La₂Ti₂O₇.

Z-scheme 2D/2D photocatalysts (Fig. 9c) can not only hold the attractive 2D/2D contact mode but also benefit from the Z-scheme photocatalytic system which can attain higher redox capacities than the traditional heterojunction [145–149]. She *et al.* [150] synthesized an α-Fe₂O₃/g-C₃N₄ 2D/2D Z-scheme hybrid photocatalyst, in which the CB electrons of α-Fe₂O₃ could easily jump to the VB of g-C₃N₄ and recombine with the photoinduced holes in the VB of g-C₃N₄ (Fig. 11a). As a result, the electron-hole recombination in both α-Fe₂O₃ and g-C₃N₄ were dramatically suppressed and the 2D/2D tight interface could pave the efficient transfer of the photoexcited electron to the reactant. Under visible light irradiation, the α-Fe₂O₃/g-C₃N₄ heterojunction displayed a very higher hydrogen production rate of 31,400 μmol g⁻¹ h⁻¹ (Fig. 11b). Soon afterwards, Xu and coworkers [92] also prepared an α-Fe₂O₃/g-C₃N₄ 2D/2D heterojunction photocatalyst, again confirming the Z-scheme charge transfer route and 2D/2D structure advantages for efficient photocatalytic H₂ evolution. Another g-C₃N₄-based 2D/2D Z-scheme photocatalyst was fabricated by incorporating hydrogen-treated WO₃ nanosheets (HWO) to Pt-loaded g-C₃N₄ nanosheets (Pt-CN) [151], and used for efficiently catalyzing the H₂ generation reaction. Similarly, 2D WO₃ nanosheets were also employed to couple with some 2D chalcogenides to form 2D/2D Z-

Figure 11 (a) Z-scheme mechanism in $\alpha\text{-Fe}_2\text{O}_3/\text{g-C}_3\text{N}_4$ hybrids. (b) Photocatalytic H₂ evolution over $\alpha\text{-Fe}_2\text{O}_3/\text{g-C}_3\text{N}_4$ hybrids. Reprinted with permission from Ref. [150]. Copyright 2017, Wiley-VCH. (c) Schematic diagrams of mechanisms for photocatalytic H₂ evolution over WO₃/ZnIn₂S₄ samples. Reprinted with permission from Ref. [90]. Copyright 2019, Royal Society of Chemistry. (d) Schematic of the photocatalytic water splitting over the 2D/2D Cu₂S/Zn_{0.67}Cd_{0.33}S. Reprinted with permission from Ref. [35]. Copyright 2019, Royal Society of Chemistry. (e) Z-scheme mechanism in BP/Bi₂WO₆ hybrids. Reprinted with permission from Ref. [153]. Copyright 2019, Wiley-VCH.

scheme photocatalysts. Huang *et al.* [152] reported the combination of CdS nanosheets or CdS/rGO nanosheets with ultrathin WO₃ nanosheets to fabricate 2D/2D Z-scheme photocatalytic systems. In water-ethanol mixtures, the as-synthesized CdS/WO₃ and CdS/rGO-WO₃ composites demonstrated higher photocatalytic hydrogen evolution activities than pure ultrathin 2D CdS nanosheets. In another study, Tan and coworkers [90] used 2D WO₃ nanosheets to couple with a ternary 2D chal-

cogenide to prepare a Z-scheme WO₃/ZnIn₂S₄ 2D/2D composite (Fig. 11c), which exhibited a high H₂ production rate of 2,202.9 μmol g⁻¹ h⁻¹. Combining different 2D chalcogenides can also be used to form the 2D/2D Z-scheme photocatalyst. A representative example was recently reported by Shi *et al.* [35], who fabricated a Cu₂S/Zn_{0.67}Cd_{0.33}S 2D/2D atomic-level heterojunction with a lamellar hexagonal morphology. Because of the 2D/2D atomic-level compact interface and efficient Z-

scheme electron-hole separation and transport (Fig. 11d), the $\text{Cu}_2\text{S}/\text{Zn}_{0.67}\text{Cd}_{0.33}\text{S}$ photocatalyst displayed a remarkable photocatalytic hydrogen production activity ($15.27 \text{ mmol h}^{-1} \text{ g}^{-1}$). More recently, BP, a new layered 2D material, was also chosen to combine with other 2D materials to form the 2D/2D Z-scheme photocatalyst. Hu *et al.* [153] reported the assembling of 2D monolayer Bi_2WO_6 (MBWO) with 2D layered BP to form a novel Z-scheme 2D/2D heterojunction (Fig. 11e). The H_2 evolution rate of BP/MBWO can reach $21,042 \mu\text{mol g}^{-1}$, which is 9.15 times that of pristine MBWO.

More recently, a new step-scheme (S-scheme) heterojunction concept was proposed based on the Z-scheme photocatalysts [116,154–158]. As shown in Fig. 9d, two n-type semiconductor photocatalysts were coupled together to form the S-scheme heterojunction, in which one acts as an oxidation photocatalyst (Semiconductor A) and the other is reduction photocatalyst (Semiconductor B). Due to the driving force of the internal electric field in S-scheme heterojunction, the excited electrons in the CB of oxidation photocatalysts will easily recombine with holes in the VB of the reduction photocatalysts. Significantly, the strongly oxidative holes in the VB of oxidation pho-

tocatalysts and the strongly reductive electrons in the CB of reduction photocatalysts will be spatially separated. The S-scheme strategy can not only efficiently restrain the recombination of photogenerated charges but also promote the reducing capacity and oxidizing ability of the heterojunction in the photocatalysis process. A recent study reported the design of a 2D/2D $\text{WO}_3/\text{g-C}_3\text{N}_4$ S-scheme heterojunction for photocatalytic H_2 production [116], in which 2D $\text{g-C}_3\text{N}_4$ served as a reduction-type photocatalyst while 2D WO_3 was an oxidation-type photocatalyst. As shown in Fig. 12a and b, there is a difference of work function between WO_3 and $\text{g-C}_3\text{N}_4$, which indicates the presence of charge transfer at the interface of WO_3 and $\text{g-C}_3\text{N}_4$. According to the S-scheme charge transfer mechanism, the relatively useless electrons in the CB of WO_3 and holes in the VB of $\text{g-C}_3\text{N}_4$ would recombine together, whereas the useful electrons and holes in the CB of $\text{g-C}_3\text{N}_4$ and VB of WO_3 would be respectively left to participate in photocatalytic reactions (Fig. 12c–e). As a result, the electrons left in the CB of $\text{g-C}_3\text{N}_4$ exhibited supreme redox capacity, thus providing a strong driving force for running the photocatalytic water splitting reaction. In a more recent study,

Figure 12 (a) Electrostatic potentials of (a) WO_3 (001) surface and (b) $\text{g-C}_3\text{N}_4$ (001) surface. (c–e) S-scheme charge transfer mechanism between WO_3 and $\text{g-C}_3\text{N}_4$. Reprinted with permission from Ref. [116]. Copyright 2019, Elsevier.

Ren *et al.* [159] employed 2D CdS to combine with 2D g-C₃N₄ to form an S-scheme heterojunction for photocatalytic H₂ production. They also demonstrated that the formation of the 2D/2D S-scheme heterojunction could accelerate the interfacial charge separation for surface reaction. The CdS/g-C₃N₄ 2D/2D S-scheme heterojunction showed a high H₂ production rate of 153,000 $\mu\text{mol g}^{-1} \text{h}^{-1}$, which was 3.83 times and 3060 times higher than those of pure CdS and g-C₃N₄, respectively.

Another type of 2D/2D heterojunction used for hydrogen generation was formed by coupling 2D cocatalyst with 2D semiconductor photocatalysts (Fig. 9e). It has been demonstrated that suitable cocatalysts play pivotal roles in improving both the activity and reliability of semiconductor photocatalysts. The cocatalysts can not only decrease the overpotential for hydrogen production but also boost the electron-hole separation at the cocatalyst/semiconductor interface. Previously, some 0D noble metal nanoparticles were often employed as efficient cocatalysts. However, the small contact area between the 0D noble metal and 2D semiconductor severely hinders the charge transfer at the interface. Moreover, the high cost and scarcity of noble metals limit their practical applications. In recent years, various earth-abundant 2D noble-metal-free cocatalysts have been emerging as potential candidates to replace noble metals. For example, MoS₂, a rising star in 2D layered materials, has been reported as an efficient cocatalyst on a variety of 2D semiconductor photocatalysts. Yuan and coworkers [40] loaded 2D MoS₂ nanosheets on the surface of 2D anatase TiO₂ nanosheets with exposed (001) facets. The photocatalytic H₂-production activity of TiO₂ was significantly improved by loading an optimal amount of 0.50 wt% MoS₂ as a cocatalyst. Later, the authors also used 2D MoS₂ nanosheets cocatalyst to modify Cu²⁺-doped ZnIn₂S₄ (Cu-ZnIn₂S₄) nanosheets for highly efficient solar-to-H₂ conversion [160]. It was found that the 2D/2D MoS₂/Cu-ZnIn₂S₄ photocatalyst at a 6 wt% MoS₂ loading amount achieved a higher H₂-evolution rate of 5,463 $\mu\text{mol g}^{-1} \text{h}^{-1}$. Ma *et al.* [86] demonstrated that the photocatalytic H₂-evolution activity of 2D CdS nanosheets could be significantly boosted by loading ultra-thin MoS₂ nanosheets. The photocatalytic H₂ evolution performance of 2D layered niobate oxides (e.g., SnNb₂O₆) can also be improved by coupling 2D MoS₂ nanosheets [70]. More recently, Yuan's group [82] further employed 2D MoS₂ nanosheets to combine with 2D metal-free semiconductors to form BP/MoS₂ and g-C₃N₄/MoS₂ [81] 2D/2D composites. In the BP/MoS₂ 2D/2D photocatalyst

system, the MoS₂ acts as both an electron sink and a cocatalyst which will greatly reduce the recombination of electron-hole pairs, while the 2D/2D smart structure provides large 2D nanointerfaces for photogenerated charge transfer, and thus synergistically promotes the H₂ evolution rate (Fig. 13a). As a result, the BP/MoS₂ 2D/2D photocatalyst loaded with 10% MoS₂ showed a H₂ evolution rate of 1,286 $\mu\text{mol g}^{-1} \text{h}^{-1}$, which is higher than that of other counterparts (Fig. 13b). Besides MoS₂, MXene, a new family of 2D material, possesses excellent electrical conductivity, good hydrophilicity, and lower Fermi level compared with semiconductors, which could also serve as an effective cocatalyst for photocatalytic hydrogen production from water. Su *et al.* [119] synthesized 2D/2D Ti₃C₂/g-C₃N₄ composites as photocatalysts for hydrogen evolution under visible light irradiation. As shown in Fig. 13c, a Schottky barrier was present at the Ti₃C₂/g-C₃N₄ interface when Ti₃C₂ was coupled with g-C₃N₄, which can serve as the electron reservoir, thus prompting the separation of photoinduced electrons and holes. In addition, the large intimate 2D/2D interface between the g-C₃N₄ and Ti₃C₂ can shorten the charge transfer distance, leading to the greatly improved migration rate of the photoinduced electrons. The results revealed that the 2D/2D Ti₃C₂/g-C₃N₄ composites displayed a 10 times higher photocatalytic hydrogen evolution activity than that of pristine g-C₃N₄ (Fig. 13d). Similarly, Lin *et al.* [118] fabricated a 2D/2D Ti₃C₂ MXene/O-doped g-C₃N₄ Schottky-junction photocatalyst, which exhibited a high hydrogen evolution rate of 25,124 $\mu\text{mol g}^{-1} \text{h}^{-1}$.

Multicomponent 2D cocatalysts have also been investigated to improve the photocatalytic hydrogen production performance of semiconductor photocatalysts. Li *et al.* [75] reported that 2D TiO₂ with co-exposed (101) and (001) facets was modified by Ti₃C₂ and MoS₂ bi-component cocatalysts toward enhanced photocatalytic hydrogen production activity. In this ternary 2D/2D/2D Ti₃C₂@TiO₂@MoS₂, the photoexcited electrons can migrate from the (001) facets of TiO₂ to (101) facets and Ti₃C₂, respectively, and then the electrons on the (101) facets transport to MoS₂, leading to a dual-carrier-separation manner (Fig. 13e). Therefore, an electron-rich environment was acquired on the planar surfaces of Ti₃C₂ and MoS₂, on which the H₂O was reduced to produce H₂ with a higher rate of 6,425.297 $\mu\text{mol h}^{-1} \text{g}^{-1}$ (Fig. 13f). In some other studies, other 2D sulfide-based materials as well metal phosphides were also employed as cocatalysts to modify some 2D semiconductor photocatalysts for boosting the performance of photocatalytic hydrogen evolution, such as Ni₂P/ZnIn₂S₄ [110] and SnS₂/TiO₂ [78].

Figure 13 Schematic diagrams of the composite photocatalysts and the corresponding charge transfer route, and comparison of the H₂ production rates over the composite photocatalysts with their respective counterparts. (a, b) 2D/2D BP/MoS₂. Reprinted with permission from Ref. [82]. Copyright 2019, Elsevier. (c, d) 2D/2D Ti₃C₂/g-C₃N₄. Reprinted with permission from Ref. [119]. Copyright 2019, Elsevier. (e, f) 2D/2D/2D Ti₃C₂@TiO₂@MoS₂. Reprinted with permission from Ref. [75]. Copyright 2019, Elsevier.

CO₂ reduction

Nowadays, the continuous consumption of fossil fuel for industrial manufacturing has caused a terrible growing amount of CO₂ in the atmosphere [159]. It has been reported that the annual anthropogenic CO₂ emissions from fossil fuel combustion have reached at about 9 Gt (1 G=10⁹), which is approximately 43% higher than the level recorded in pre-industrial times [161,162]. The substantial rise of CO₂ in atmosphere would seriously damage the balance of the earth's carbon cycle and trigger global warming. Thus, many strategies have been explored to reduce the CO₂ concentration in atmosphere, such as electrochemical [163], biological [164], thermochemical [165] and photocatalytic means [166,167]. Among them, photocatalytically converting CO₂ into valuable energy fuels (e.g., CH₄, CH₃OH, CO) by utilizing solar energy has been regarded as a “kill two birds with one stone” ideal approach in terms of protecting our environment and simultaneously supplying energy [161,168]. Since the first discovery of photoconversion of

CO₂ to valuable fuels over semiconductor materials by Inoue and coworkers [169] in 1979, numerous studies have been conducted on the preparation of highly efficient photocatalysts to meet the requirements of practical application of CO₂ photoreduction [131,170–176]. As mentioned above, various products can be derived from photocatalytic CO₂ reduction *via* multi-step reaction pathways. The following equations give the half reactions to the various products commonly formed in CO₂ reduction [161]. In addition, the theoretical reduction potential (E^0) (V, vs. NHE at pH 7) for each half equation is also provided.

Clearly, the final products depend upon the thermodynamical mechanism to conduct the reaction, such as the number of electrons participating in the reaction, electron migration rate, and CB and VB potentials of the photocatalysts. Thus, the studies in this area need not only to obtain good efficiency but also to increase the selectivity for specific products. Particularly, the utilization of 2D/2D heterojunction nanomaterials in the field of CO_2 photoreduction has fascinated many researchers (Table 3). Combining different 2D materials can establish a strong interface contact as well as large contact surface to allow the rapid transfer and separation of photoinduced charge carriers across the heterojunction interface, thus leading to greatly improved photocatalytic CO_2 reduction efficiency and selectivity. Ong *et al.* [96] reported the incorporation of rGO with protonated g- C_3N_4 (pCN) to form an rGO-pCN 2D/2D hybrid heterojunction towards photocatalytic reduction of CO_2 to CH_4 . It has been confirmed that rGO has excellent charge mobility and high electron storage capacity, which often acts as cocatalyst to provide conductive electron channels for the separation of photogenerated charges in the heterojunction photocatalyst. In this work, the photogenerated electrons in the CB of pCN would easily migrate to rGO framework owing to the unique properties of rGO as well as the large interface contact area between pCN and rGO. Then, the photoinduced holes left in the VB of pCN will react with H_2O molecules to produce protons (H^+), while the electrons on the rGO would not only interact with the H^+ to generate $\cdot\text{H}$ radicals but also react with the activated CO_2 molecules to form superoxide ($\cdot\text{CO}_2^-$) radicals. Subsequently, the CH_4 product was obtained through a series of radical reactions between $\cdot\text{CO}_2^-$ and $\cdot\text{H}$. The results revealed that the total amount of CH_4 production was 5.4-folds higher than that of pure pCN photocatalysts. 2D rGO cocatalyst was also reported to combine with SiC nanosheets by Han and coworkers [33] for CO_2 photoreduction with high efficiency and CH_4 selectivity. In this photoreduction system, the robust 2D/2D SiC/rGO heterojunction allowed fast transfer of energetic electrons from SiC to rGO and the proportion of rGO had important effects on both the activity and selectivity. The results demonstrated that a low proportion of rGO in the composite would accumulate dense energetic electrons (Fig. 14a and b), promoting the eight-electron process for CH_4 generation, whereas a high proportion of

rGO would result in sparse energetic electrons on rGO, paving the two-electron process for CO generation. In another study, Cao *et al.* [28] employed ultrathin Ti_3C_2 nanosheets as a cocatalyst to couple with Bi_2WO_6 nanosheets for photocatalytic CO_2 reduction. Benefiting from the unique 2D/2D heterojunction, the CO_2 adsorption capability was enhanced and the photoinduced electrons could quickly transfer from Bi_2WO_6 to the surface of Ti_3C_2 (Fig. 14c). As a result, the 2D/2D $\text{Ti}_3\text{C}_2/\text{Bi}_2\text{WO}_6$ photocatalyst exhibited 4 and 6 times higher productive rates of CH_4 and CH_3OH than that of pristine Bi_2WO_6 nanosheets, respectively.

2D/2D Z-scheme photocatalysts have also been reported for enhanced photoreduction CO_2 activity. Wang *et al.* [47] synthesized Z-scheme $\text{BiOI}/\text{g-C}_3\text{N}_4$ 2D/2D photocatalyst for the reduction of CO_2 to produce CO, H_2 and/or CH_4 . Under visible light irradiation, the synthesized photocatalyst exhibited more highly efficient photoreduction CO_2 activity than pure g- C_3N_4 and BiOI along. Another example of 2D/2D hybrid Z-scheme heterojunction used for photoreduction of CO_2 was reported by Jo and coworkers [50]. In this work, they constructed a $\text{Bi}_2\text{WO}_6/\text{rGO}/\text{g-C}_3\text{N}_4$ hybrid heterojunction, in which the rGO acted as not only a supporter to capture the electrons from g- C_3N_4 , but also the redox mediator to promote the Z-scheme charge transfer be-

Figure 14 (a, b) Schematic diagrams of high (a) and low (b) electron density-dependent CH_4 selectivity in SiC/rGO heterojunctions. Reprinted with permission from Ref. [33]. Copyright 2018, Wiley-VCH. (c) Energy level structure diagram of Bi_2WO_6 and Ti_3C_2 , as well as the photoinduced electron transfer process. Reprinted with permission from Ref. [28]. Copyright 2018, Wiley-VCH.

tween g-C₃N₄ and Bi₂WO₆ (Fig. 15b). Profiting from the effects of Z-scheme charge transfer route, large interfacial contact of the 2D/2D architecture and the advantages of rGO, the Bi₂WO₆/rGO/g-C₃N₄ hybrid photocatalyst showed a higher yield of carbonaceous products (CO + CH₄) as well as enhanced selectivity of the products (Fig. 15a).

The 2D/2D heterojunction photocatalysts with a Type-II configuration have also been applied in photocatalytic CO₂ reduction. Tonda *et al.* [57] constructed a 2D/2D Type-II heterojunction between the negatively charged

2D g-C₃N₄ nanosheets and positively charged NiAl-LDH for photocatalytic CO₂ reduction. Because of the synergistic effect of 2D/2D strong interfacial contact and efficient charge carrier transfer mode of Type-II, the resulting 2D/2D g-C₃N₄/NiAl-LDH showed 5- and 9-fold enhancement of CO production rate than pure g-C₃N₄ and NiAl-LDH, respectively. More recently, Kong and coworkers [125] developed 2D/2D surface defect-engineered Bi₂WO₆/BiOI p-n heterojunction photocatalysts (BWO-OV/BOI) towards superior photocatalytic CO₂ reduction activity. As show in Fig. 15c, the authors

Figure 15 (a) Comparison of the photocatalytic CO, CH₄, H₂, and O₂ production rates over the Bi₂WO₆/rGO/g-C₃N₄ samples as well as other synthesized photocatalysts. (b) Schematic illustration of the proposed mechanism for CO₂ photoreduction in the Bi₂WO₆/rGO/g-C₃N₄ sample. Reprinted with permission from Ref. [50]. Copyright 2018, Elsevier. (c) Estimated relative band positions and schematic diagram of charge transfer and separation at the BWO-OV/BOI composite. (d) Schematic illustration for the proposed mechanism of photocatalytic CO₂ reduction over the BWO-OV/BOI composite. (e) Total yield of CH₄ production over the BWO-OV/BOI composite and other as-developed samples. (f) Photostability tests of BWO-OV/BOI composite. Reprinted with permission from Ref. [125]. Copyright 2019, Elsevier.

pointed out that the migration of charge carriers in this system was through the p-n heterojunction with the built-in internal electric field instead of the conventional Type-II heterojunction when Bi_2WO_6 and BiOI came into contact. What is more, the 2D/2D motif allowed directional migration and spatial separation of photoinduced charge carriers to opposite sides in the heterostructure, which effectively hindered their recombination (Fig. 15d). The BWO-OV/BOI photocatalytic system displayed a remarkable visible-light-driven CH_4 production yield of $18.32 \mu\text{mol g}^{-1}$, which is 7.1-fold enhancement over the pure BOI photocatalysts (Fig. 15d). In addition, the CH_4 production yield over the BWO-OV/BOI composite can be maintained about 92% of the initial value after three consecutive test cycles, suggesting its superior photostability (Fig. 15e).

Removal of pollutions

In the past few decades, the rapid development of urbanization and industrialization has simultaneously led to the increasing serious environmental problems, such as discharge of wastewater and spoil gas that often contain various toxic species, which do great harm to human health and ecosystem equilibrium [177,178]. Thus, a great number of methods, including chemical degradation [179], physical adsorption [180], and biodegradation techniques [181] have been adopted for environment remediation. However, most of the conventional treatments for environment remediation were found inefficient and may cause secondary pollution. In recent years, sunlight-driven photocatalysis technology is considered to be a green, effective and economic method to handle the removal of environmental pollutions [182–187]. To date, tremendous efforts have been contributed to developing efficient photocatalysts to match the requirements of practical application [188–191]. It is well accepted that the development of heterostructured photocatalysts comprising multiple components possess efficient charge carrier separation abilities as compared with that of the single one, which is deemed as a promising strategy for achieving highly efficient photocatalytic activity towards environment remediation [192–194]. In particular, the composite photocatalysts with 2D/2D heterojunction often display more excellent separation of electron-hole pairs due to the intimate face-to-face contact, leading to the enhanced photocatalytic removal of environmental pollutions. Up to now, plentiful of 2D/2D heterojunctions have been applied for the removal of various kinds of pollutions, which are summarized in Table 3. Among many 2D/2D materials, the $\text{g-C}_3\text{N}_4$ based

heterojunctions are the most frequently studied photocatalysts applied in environmental remediation. For example, 2D chalcogenide was often coupled with $\text{g-C}_3\text{N}_4$ to form a 2D/2D heterojunction towards the elimination of environmental pollution. Zhang *et al.* [97] fabricated 2D/2D $\text{SnS}_2/\text{g-C}_3\text{N}_4$ heterojunction nanosheets for the degradation of organic dyes and phenols. The formed Type-II heterojunction between SnS_2 and $\text{g-C}_3\text{N}_4$ as well as the larger contact interface region of 2D/2D construction greatly improved the separation of the electron-hole pairs, thus resulting in the enhanced photocatalytic activities for the degradation of pollutants as compared with pure $\text{g-C}_3\text{N}_4$ and SnS_2 nanosheets. More recently, Huo *et al.* [69] and Song *et al.* [123] further studied the uses of 2D/2D $\text{SnS}_2/\text{g-C}_3\text{N}_4$ heterojunction for photodegradation of organic dyes, respectively. However, both of the authors of these two studies pointed out that charge migration mechanism between SnS_2 and $\text{g-C}_3\text{N}_4$ should be a Z-scheme mode rather than the conventional Type-II. The $\text{MoS}_2/\text{g-C}_3\text{N}_4$ 2D/2D heterojunction photocatalysts were also developed to remove environmental pollutants [100,101]. Some 2D ternary chalcogenide semiconductors were also used to couple with $\text{g-C}_3\text{N}_4$ to form 2D/2D heterojunction photocatalysts for degradation of pollutants.

Guo *et al.* [68] developed a 2D/2D $\text{CuInS}_2/\text{g-C}_3\text{N}_4$ Z-scheme heterojunction towards visible-light-driven photocatalytic degradation of tetracycline (TC). The results revealed that the $\text{CuInS}_2/\text{g-C}_3\text{N}_4$ heterojunction showed a higher apparent degradation rate than that of pure $\text{g-C}_3\text{N}_4$ and CuInS_2 nanosheets. Similarly, 2D MnInS_4 semiconductor was also reported to combine with $\text{g-C}_3\text{N}_4$ to construct Z-scheme 2D/2D architectures for treatment of pharmaceutical wastewater [66]. In another study, Che *et al.* [195] employed yeast-derived carbon (YC) spheres as a charge carrier bridge to the 2D/2D $\text{Cu}_2\text{WS}_4/\text{g-C}_3\text{N}_4$ heterojunction towards photocatalytic reduction of hexavalent chromium Cr(VI) and decomposition of TC. The $\text{g-C}_3\text{N}_4/\text{YC}/\text{Cu}_2\text{WS}_4$ heterojunction congregated the merits of face-to-face 2D/2D architectures, introducing a bridge for electron migration and Type-II charge transfer mode, which significantly inhibit the rapid recombination of charge carriers (Fig. 16a). As a result, The $\text{g-C}_3\text{N}_4/\text{YC}/\text{Cu}_2\text{WS}_4$ showed enhanced photocatalytic activity for decomposing TC (Fig. 16b) and Cr(VI) reduction. Some bismuth-based semiconductors with a layered structure are also incorporated with $\text{g-C}_3\text{N}_4$ to get the 2D/2D heterostructured composite toward degradation of contaminant. Wang *et al.* [196] constructed a novel atomic scale 2D/2D $\text{g-C}_3\text{N}_4/\text{Bi}_2\text{WO}_6$ heterojunction with a Type-

Figure 16 (a) The proposed mechanism for the enhancement of photocatalytic activity over $\text{Cu}_2\text{WS}_4/\text{YC}/\text{g-C}_3\text{N}_4$ heterojunction. (b) TC degradation dynamics curves over $\text{Cu}_2\text{WS}_4/\text{YC}/\text{g-C}_3\text{N}_4$ and other counterparts. Reprinted with permission from Ref. [195]. Copyright 2019, Elsevier. (c) Photocatalytic mechanism scheme of $\text{g-C}_3\text{N}_4/\text{Bi}_2\text{WO}_6$ 2D/2D heterojunction. (d) Photocatalytic degradation of ibuprofen by using $\text{g-C}_3\text{N}_4/\text{Bi}_2\text{WO}_6$ and other samples. Reprinted with permission from Ref. [196]. Copyright 2017, Elsevier.

II band alignment (Fig. 16c), which displayed almost 96.1% ibuprofen removal efficiency of after 1 h visible light irradiation (Fig. 16d). Later, Guo and coworkers [111] studied that the Bi_2WO_6 and $\text{g-C}_3\text{N}_4$ can be constructing a Z-scheme 2D/2D heterojunction to boost photoinduced charge carriers separation, leading to an enhanced visible-light-driven photodegradation efficiency. Combining $\text{g-C}_3\text{N}_4$ with bismuth oxyhalides to form 2D/2D heterojunction also obtained much attention in the degradation of organic pollutants. Wang *et al.* [58] designed an oxygen vacancy (OV)-rich ultrathin $\text{g-C}_3\text{N}_4$ - BiOCl 2D/2D heterostructure nanosheet, which showed 95% removal efficiency of 4-chlorophenol within 2 h visible light irradiation. In another study, Che and coworkers [197] employed $\text{Bi}_3\text{O}_4\text{Cl}$ to couple with $\text{g-C}_3\text{N}_4$ to assembly Z-scheme 2D/2D heterojunctions. Under visible light irradiation, the Z-scheme $\text{Bi}_3\text{O}_4\text{Cl}/\text{g-C}_3\text{N}_4$ 2D/2D heterojunctions exhibited outstanding photocatalytic activity for removing the various water contaminants. Besides these, some other types of 2D bismuth-contained semiconductors were also reported to couple with $\text{g-C}_3\text{N}_4$ to construct 2D/2D heterojunctions for environmental remediation, such as $\text{C}_3\text{N}_4/\text{Bi}_{20}\text{TiO}_{32}$ [29], $\text{BiOIO}_3/\text{g-C}_3\text{N}_4$ [109], $\text{Bi}_2\text{O}_2\text{CO}_3/\text{g-C}_3\text{N}_4$ [99], $\text{BiVO}_4/\text{g-C}_3\text{N}_4$ [120]. It can

be found that the above discussed $\text{g-C}_3\text{N}_4$ based 2D/2D photocatalysts contained at least one metal element, which would raise economic cost concern of large-scale production. Thus, the construction of $\text{g-C}_3\text{N}_4$ based metal-free 2D/2D materials with earth-abundant elements have emerged as attractive photocatalysts for pollutant degradation in recent years. GO or rGO was often selected to combine with $\text{g-C}_3\text{N}_4$ nanosheets to form a stable 2D/2D metal-free hybrid because their excellent adsorption performance and outstanding electroconductivity. Typically, the photogenerated electrons of $\text{g-C}_3\text{N}_4$ can rapidly transfer to GO or rGO *via* the percolation mechanism to inhibit the recombination of electron-hole pairs, and thereby leading to boosted photocatalytic activity for pollutant degradation [98,198,199]. Typically, the photogenerated electrons of $\text{g-C}_3\text{N}_4$ can rapidly transfer to GO or rGO *via* the percolation mechanism to inhibit the recombination of electron-hole pairs, and thereby leading to boosted photocatalytic activity for pollutant degradation. Another type of metal-free 2D/2D photocatalysts was composed of BP and $\text{g-C}_3\text{N}_4$. In the previous section, we have discussed that the BP/ $\text{g-C}_3\text{N}_4$ 2D/2D heterojunction can be applied in photocatalytic hydrogen production. The BP/ $\text{g-C}_3\text{N}_4$ 2D/

2D photocatalysts could also be used to photodegrade organic pollutants. Wang's group [200] demonstrated that the BP/g-C₃N₄ photocatalysts showed better photocatalytic activity in degradation of rhodamine B (RhB) under visible-light irradiation. More recently, Zhang *et al.* [124] employed the BP/g-C₃N₄ photocatalysts for photocatalytic degradation of bisphenol A (BPA). The BPA degradation rate can reach to 88% over this dual-function photocatalytic system.

Bismuth-contained 2D/2D photocatalysts is another category that is often used for environmental remediation. In addition to the above discussed in coupling with g-C₃N₄, the bismuth-based 2D layer materials can also be hybridized with other 2D photocatalysts to form 2D/2D heterojunction for the degradation of pollutants. Sultana *et al.* [107] reported a composite of 2D-CeO₂ with 2D BiOI to form a Z-scheme heterojunction for RhB dye decolorization and phenol degradation. In another study, Xu's group [91] designed Au nanoparticles (NPs)-decorated

2D/2D Bi₂WO₆-TiO₂ heterostructure (Fig. 17a) for photocatalytic reduction of nitroaromatics and heavy metal ions Cr(VI). In this hybrid photocatalyst system, the cooperative synergy effect (Fig. 17b), including Z-scheme as well as 2D/2D rapid charge transfer platforms and surface plasmon resonance effect as well as the "electron sink" by decorated Au NPs, resulted in a boosted photocatalytic performance. More recently, a 2D/2D S-scheme In₂S₃/Bi₂O₂CO₃ heterojunction photocatalyst was also designed for RhB degradation [156]. Benefiting from the S-scheme charge transfer mechanism as well as the face-to-face 2D/2D structure (Fig. 17c), the 2D/2D In₂S₃/Bi₂O₂CO₃ heterojunction showed 5 and 3 times RhB degradation capacity higher than that of pure Bi₂O₂CO₃ and In₂S₃ (Fig. 17d). 2D perovskite type semiconductor was also used to combine with bismuth-contained photocatalysts towards pollutant destruction. Ao *et al.* [45] reported the coupling perovskite type semiconductor lanthanide titanate (La₂Ti₂O₇) with bismuth oxybromide (BiOBr) to

Figure 17 (a) TEM image of Au NPs decorated 2D/2D Bi₂WO₆-TiO₂ heterostructure. (b) The proposed mechanism for the enhancement of photocatalytic activity over Au NPs decorated 2D/2D Bi₂WO₆-TiO₂ heterostructure. SPR: surface plasmon resonance. Reprinted with permission from Ref. [91]. Copyright 2017, Wiley-VCH. (c) Sketch of the 2D/2D In₂S₃/Bi₂O₂CO₃ S-scheme heterojunction. (d) Photocatalytic degradation rates over the In₂S₃/Bi₂O₂CO₃ and other fabricated samples. Reprinted with permission from Ref. [156]. Copyright 2020, Elsevier.

form 2D/2D p-n type heterojunctions. Under visible light irradiation, the formed BiOBr/La₂Ti₂O₇ p-n heterojunction showed an enhanced photocatalytic degradation activity to dye RhB and phenol. Later, the authors in the same group [46] further synthesized BiOCl/La₂Ti₂O₇ 2D/2D p-n type heterojunction for photocatalytic degradation of RhB. Two different types of bismuth-contained semiconductors also can be combined to realize the 2D/2D photocatalyst system for environmental remediation, such as BiOI₃/BiOI [41], Bi₂S₃-BiOCl [201], Bi₄Ti₃O₁₂/I-BiOCl [31] and BiOI/BiVO₄ [55].

Besides the aforementioned g-C₃N₄ based and bismuth-containing photocatalysts, there are also other advanced 2D/2D heterojunctions used for environmental remediation, such as SnNb₂O₆/graphene [95], WO₃/SnNb₂O₆ [108], WC/WO₃ [202] and TiO₂/WS₂ [79].

CONCLUSION AND OUTLOOK

In the past few years, significant progresses have been made in the synthesis and applications of 2D/2D heterostructures. The 2D/2D heterostructures possess many superior properties, such as integrating the merits of each 2D component and dramatically enhanced separation or transfer of charges. Driven by these advantages, 2D/2D heterostructures were deemed as excellent candidates for fundamental photocatalytic research and potential commercial applications. In this comprehensive review paper, we have highlighted the advanced progress of 2D/2D heterostructure photocatalysts from their general designing strategies to representative photocatalytic applications. As demonstrated by a large amount of literature, the rational design of 2D/2D heterostructures can significantly boost their photocatalytic activities, mainly attributed to their intrinsic 2D lamellar nature, large contact area and strong interactions. Till now, some representative 2D/2D heterostructures, such as g-C₃N₄/MoS₂, BP/g-C₃N₄, BP/MoS₂, Ti₃C₂/MoS₂, α -Fe₂O₃/GO and g-C₃N₄/rGO, have shown significant enhancement of photocatalytic performance for hydrogen evolution, CO₂ reduction and degradation of pollutants as compared with pristine 2D materials.

Despite considerable achievements in the design and application of 2D/2D heterostructure photocatalysts, there are still several challenges ahead in this area. In the first aspect, it is well-known that the large quantity manufacture of photocatalyst materials is of great importance for the potential commercial applications. However, there are few scalable strategies to produce the 2D/2D heterostructures in large scale with controllable construction. Thus, more effort should be concentrated

on the development of low-cost and large-scale production approaches. The second obstacle is the serious agglomeration issue when the different 2D components were coupled together, which would result in loss of the unique structural benefits of the 2D morphology. In this regard, it encourages developing some strategies to overcome the surface energies of the 2D/2D heterostructures for better stabilization of freestanding in the 2D architecture. The third challenge is the lack of study on the effect of the thickness of each 2D layer on the performances of 2D/2D heterostructures. Theoretically, the photocatalytic performances of the 2D materials are highly dependent on their thickness. For example, the atomic thickness of ultrathin 2D nanomaterials can endow them with high specific surface area, ultimate exposure of their surface atoms and excellent optical transparency, making them exceedingly desirable in the application of photocatalysis. In addition, the thickness of 2D nanomaterials also affects their electronic band structure, which plays a vital role in photocatalytic the application. However, fewer studies have been conducted to investigate the relationship between the thickness of 2D materials and the photocatalytic performances of their 2D/2D heterostructures. In view of this point, the relationship between the thicknesses and the photocatalytic performances of 2D/2D heterostructures can be investigated by controlling the thickness of each 2D layer. The fourth aspect is a huge lack of deep understanding of the physical and chemical properties as well as fundamental formation mechanisms of the 2D/2D heterostructure photocatalysts. In this regard, some direct and accurate characterization techniques can be adopted to uncover the intrinsic feature and the real charge transfer pathway over the 2D/2D heterostructures during photocatalytic reactions, such as *in-situ* Raman spectroscopy, *in-situ* XPS and *in situ* electron microscopy. In addition, the rational fundamental calculations and simulations based on density functional theory (DFT) should be also paid much more attention to, because they cannot only provide a better fundamental understanding of the formation as well photocatalytic mechanism but also guide the design of efficient 2D/2D photocatalysts. Finally, while the researchers explore novel 2D/2D photocatalysts to meet the requirements of highly efficient photocatalytic performance, one should also pay some attention to that there is plenty of space for developing the existing 2D/2D heterostructures for potential photocatalytic applications, e.g., hydrogen peroxide production [240].

Benefiting from the rapid development and abundant knowledge accumulated in 2D materials as well as their

heterostructures in recent years, one may expect that 2D/2D hybrid photocatalysts would play an important role in solving energy and environmental crisis. It is believed that this comprehensive review will contribute to the further research in the area of 2D/2D materials or photocatalysis.

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Conflict of interest The authors declare that they have no conflict of interest.

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二维/二维异质结光催化剂：合理设计与能源和环境应用

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摘要 近年来, 二维/二维异质结纳米材料在光催化领域引起了广泛的研究. 二维/二维异质结材料在高效光催化剂的应用研究上具有许多独特优点, 主要包括良好的尺寸设计, 较大的界面接触面积, 有机集合了各二维组分的优点, 同时异质结的构筑能够快速促进光生电荷的分离和转移. 本文首先介绍了二维/二维异质结构光催化剂所形成界面的一些基本原理内容, 并总结了目前二维/二维异质结构光催化剂的一般合成策略, 包括原位生长和非原位外组装. 随后着重介绍了二维/二维异质结光催化剂在产氢、二氧化碳还原和污染物去除等方面的应用的最新研究进展. 最后, 对二维/二维异质结光催化剂的发展前景进行了展望.