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SPECIAL TOPIC: Heterojunction in Photocatalysts

g-C₃N₄-based S-scheme heterojunction photocatalysts

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ABSTRACT With the vigorous progress of industrialization, energy shortage and environmental contamination emerge increasingly serious. Photocatalysis technology is known as a hopeful approach to resolving the above crises owing to its numerous prominent advantages and widespread applications. Among various photocatalysts, graphitic carbon nitride $(g-C_3N_4)$ has been broadly applied in fields of fuel production and environment purification because of its unique electronic structure, extreme thermal stability, and prominent photoelectrical activity. However, single-component g-C₃N₄, similar to other photocatalysts, usually suffer from low photocatalytic efficiency due to the fact that single-constituent photocatalysts cannot synchronously equip with strong redox abilities of photogenerated charges and high light energy utilization. Fortunately, constructing Step-scheme (S-scheme) heterojunctions between g-C₃N₄ with other semiconductor photocan simultaneously overcome the typical catalysts shortcomings of low light energy utilization, rapid recombination, and weak redox abilities of carriers, thus prominently boosting its catalytic reaction rate. In view of the currently extensive reports of g-C₃N₄-based S-scheme heterojunctions, this review presents a relatively comprehensive comment on the latest research progress of the background, the proposal of conception, fundamental theory, design and preparation, characterization methods of g-C₃N₄-based S-scheme heterojunctions. Additionally, various applications of g-C₃N₄-based S-scheme heterojunctions have been detailly illustrated through example discussion and list comparison, involving photocatalytic H₂ generation, CO₂ reduction, H₂O₂ evolution, pollutant degradation, and others. Finally, the research progress and shortcomings of g-C₃N₄-based S-scheme heterojunctions are summarized, and the future research direction is prospected.

Keywords: photocatalysis, g-C₃N₄, heterojunction, S-scheme, applications

INTRODUCTION

With the vigorous progress of industrialization, energy shortage and environmental contamination emerge increasingly serious [1,2]. Photocatalysis is known as a hopeful technology to resolve the two major energy and environment crises because of its low cost, mild reaction process, and eco-friendly advantages [3–5]. In other words, photocatalysis technology refers that under the drive of solar energy, photocatalysts can trigger a series of reactions to produce fuels and repair the environment involving H_2 production, CO_2 reduction, antibiotic removal, and pollution degradation [6,7]. In the case of identical external conditions, the efficiency of photocatalysis technology is mainly depended on the photocatalysts [8,9]. Therefore, it is very necessary to develop efficient photocatalysts. Graphitic carbon nitride (g- C_3N_4), a representative of organic photocatalysts, has drawn considerable attention since its first application in H_2 evolution because of its unique electronic structure and prominent photoelectrical activity [10]. In recent years, $g-C_3N_4$ has been widely applied in the fields of H_2 production, CO_2 reduction, H_2O_2 production, and environment purification [11].

For the pure $g-C_3N_4$, similar to other single-constituent photocatalysts, usually suffers low photocatalytic efficiency owing to its low light energy utilization, rapid recombination, and weak redox abilities of photogenerated charges [12]. In this case, many strategies have been designed to overcome the above shortcomings to enhance the photocatalytic efficiency of g-C₃N₄, such as elemental doping, cocatalyst modification, and improvement of specific surface areas or crystallization degree [13,14]. However, none of the above modification strategies can simultaneously resolve the above typical shortcomings of the single photocatalyst. Namely, single-constituent photocatalysts cannot synchronously equip with strong redox abilities of photogenerated charges and high light energy utilization [15]. Additionally, the drawback of low carrier separation efficiency has always been the bottleneck hindering the improvement of photocatalysis efficiency. In view of the above-mentioned facts, constructing heterojunctions between g-C₃N₄ with another semiconductor photocatalyst can simultaneously overcome the typical shortcomings of low utilization efficiency solar energy, rapid recombination, and weak redox abilities of carriers [16], thus prominently improving the catalytic reaction of g-C₃N₄. In recent years, numerous g-C₃N₄-based heterojunctions have been constructed and aimed at enhancing the photocatalytic activity.

According to the difference in carrier transmission paths, the previously reported heterojunctions can be divided into Type-II, Z-scheme, and S-scheme, which will be briefly illustrated as follows [17]. For the Type-II heterojunction in Fig. 1a, such a carrier transfer process would not only cause the reduction of redox ability of photogenerated charges but also generate energy loss due to the repulsion between the same kind of charges [18]. To resolve the emerged deficiency in the Type-II heterojunction, a liquid Z-scheme heterojunction concept was put forward [19].

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Figure 1 Possible charges migration ways for (a) Type-II, (b) liquid Z-scheme, (c) all-solid-state Z-scheme, and (d) S-scheme heterojunctions.

In contrast to the Type-II heterojunction, the obvious difference for liquid Z-scheme heterojunction is that the suitable redox couples were added and served as the charge transport channel (Fig. 1b). In fact, there is still serval irrationality in the liquid Zscheme heterojunction. Firstly, the designed charge migration direction is not scientific because the reduction capacity of the photoinduced electrons in the Semiconductor 2 (S2) is stronger than that of Semiconductor 1 (S1), and it is much easier to reduce Fe^{3+} into Fe^{2+} (Fig. 1b). Secondly, the redox ion pairs would freely move around in the system and not be exactly fixed between the two semiconductors. Hence, an all-solid-state Zscheme concept was proposed with solid metal conductors instead of liquid redox ion pairs (Fig. 1c) [20], which could effectively avoid the problem of random movement of ion pairs in liquid Z-scheme. However, the proposed all-solid-state Zscheme still has the same unscientific carrier migration direction as the liquid Z-scheme. Consequently, both the Type-II and Zscheme heterojunction systems exhibit some unscientific and illogical aspects, which would promote the birth of a fire-new scientific and rational heterojunction concept [21].

In 2019, a fire-new concept of Step-scheme (S-scheme) heterojunction was put forward by Fu *et al.* [22]. Specifically, Yu and co-workers [23,24] constructed a $WO_3/g-C_3N_4$ heterojunction photocatalyst and scientifically explained the reason for its significantly enhanced activity by a novel S-scheme heterojunction mechanism. Since then, S-scheme heterojunction has been extensively and deeply studied by scientific research workers all over the world. In a S-scheme system, the photoexcitation charges with powerful redox capacity are retained to be involved in interfacial catalytic reaction, while the weak ones are recombined due to the function of a built-in electric field (Fig. 1d) [25]. Therefore, constructing of S-scheme heterojunction can not only strengthen the redox ability of photoexcitation carriers but also effectively promote the separation and transmission of theirs, thus greatly boosting the catalytic reaction rates of S-scheme heterojunction systems.

In view of the above prominent advantages of the S-scheme heterojunction, such as reinforced redox ability, separation efficiency, and optical absorptivity, g-C₃N₄-based S-scheme heterojunction photocatalytic systems have been widely designed to increase the photocatalytic reaction rate of g-C₃N₄. Herein, this review presents a relatively comprehensive comment on the latest research progress of g-C₃N₄-based S-scheme heterojunctions, involving the background and proposal of conception for S-scheme heterojunction. Moreover, the fundamental theory, design, and preparation, characterization methods of g-C₃N₄-based S-scheme heterojunctions are explained by theory and examples. Then, the various applications of g-C₃N₄based S-scheme heterojunctions have been detailly illustrated through example discussion and list comparison, including the photocatalytic H₂ evolution, CO₂ reduction, H₂O₂ production pollutant degradation, and others. Finally, the research progress and shortcomings of g-C₃N₄-based S-scheme heterojunctions are summarized, and the future research direction is prospected.

g-C₃N₄-BASED S-SCHEME HETEROJUNCTIONS

Fundamental theory

The S-scheme heterojunction charge transfer mechanism has been widely recognized and used by numerous scientists, mainly because of its rationality and scientificity, which is completely different from traditional Type II and Z-scheme heterojunctions. In an S-scheme heterojunction system (Fig. 2a), it is generally made up of an oxidation photocatalyst (OP) and a reduction photocatalyst (RP). When the OP and RP establish contact, the free electrons on the conductive band (CB) of RP will be driven to transfer to the CB of OP by Fermi level matching balance, thereby building an interfacial electric field from RP to OP (Fig. 2b) [21]. Moreover, the energy bands of OP will bend downward at the heterogeneous interface, while the RP will reverse. Upon light irradiation, the photoexcited holes and electrons with weak redox ability on the valence band (VB) of RP and CB of OP will occur recombination owing to the attraction of interfacial electric field, while the photogenerated charges with powerful redox capacity will be retained onto the CB of RP and VB of OP by the repulsion of interfacial electric field, respectively. Finally, the charges with powerful redox capacity will take part in the following photocatalytic reaction (Fig. 2c) [26]. Consequently, the charge transmission path of Sscheme heterojunction follows the scientific principle, and its migration mechanism not only ensures efficient carrier separation but also enhances the redox capacity of the photocatalytic system.

Design and preparation

As is well known, the conduction and valence-band potentials of semiconductors are the primary parameters to be considered for designing and preparing heterojunctions. For instance, an Sscheme heterojunction system usually involves a reduction and an OP [27]. Specifically, a semiconductor with a very negative conduction potential is commonly used as an RP, while the positive one is generally served as an OP [28]. g-C₃N₄, one of the representatives of organic polymer photocatalysts, exhibits a relatively moderate band structure and small bandgap (2.7 eV), has been broadly combined with other photocatalysts to establish heterojunctions [29]. Due to its comparatively negative reduction potential, g-C₃N₄ is generally used as an RP to construct S-scheme heterojunctions with some OPs such as WO₃, TiO₂, and Fe₂O₃ in Fig. 3 [30]. Additionally, g-C₃N₄ is also combined with some more reductive semiconductors (such as CuInS₂ and ZnCoS) to form S-scheme heterojunctions, where g-C₃N₄ acts as the OP [31]. Consequently, g-C₃N₄-based S-scheme heterojunctions have been abundantly established due to the prominent electronic structure and photoelectrical properties of g-C₃N₄.

According to the above design regulations, varieties of preparation methods have been explored to synthesize $g-C_3N_4$ based S-scheme heterojunctions, such as electrostatic selfassembly, hydrothermal, high-temperature calcination, vapour deposition, solvothermal, and thermal polymerization methods [30]. For instance, Fu *et al.* [22] constructed a two-dimensiaonal (2D)/2D WO₃/g-C₃N₄ heterojunction by an electrostatic selfassembly strategy (Fig. 4a), which involves the initial exfoliation of bulk g-C₃N₄ and WO₃ into nanosheet structure and the following surface treatment making them with positive and negative charges, respectively, resulting in the final combination of WO₃ and g-C₃N₄ nanosheets *via* Coulomb attraction. Moreover, correlation test reports suggested that the WO₃/g-C₃N₄ heterojunction formed by Coulomb force attraction had good contact, high stability, and significantly enhanced photocatalytic activity,



Figure 2 Fundamental theory of carrier-transfer for S-scheme heterojunction.



Figure 3 Band gaps and positions of some representative photocatalysts.



Figure 4 Graphic illustration of (a) $WO_3/g-C_3N_4$ heterojunctions by an electrostatic self-assembly strategy. Reprinted with permission from Ref. [22]. Copyright 2019, Elsevier. (b) $InVO_4/g-C_3N_4$ heterojunctions *via* a hydrothermal method. Reprinted with permission from Ref. [32]. Copyright 2021, Elsevier.

and its carrier migration mode followed the S-scheme heterojunction mechanism. Additionally, Gong *et al.* [32] fabricated a InVO₄/g-C₃N₄ heterojunction *via* a facile hydrothermal method, including primarily premixing the raw materials formed InVO₄ and subsequently mixing with g-C₃N₄, finally undergoing a hydrothermal treatment (Fig. 4). In fact, during the finally hydrothermal treatment, InVO₄ was *in-situ* generated onto the g-C₃N₄ surface to form the InVO₄/g-C₃N₄ S-scheme heterojunction. Similarly, the resulting InVO₄/g-C₃N₄ S-scheme heterojunction also possessed well-contact heterojunction interfaces and prominent photocatalytic activities. On the whole, most g-C₃N₄-based S-scheme heterojunctions produced by the currently reported methods have good contact and stability because of the unique interface structure and thermal polycondensation of g-C₃N₄.

Characterization method

Until now, a variety of advanced characterization techniques have been developed to investigate the charge transfer path of g-C₃N₄-based S-scheme heterojunctions, such as *in-situ* irradiated X-ray photoelectron spectroscopy (ISIXPS), density functional theory (DFT) calculation, electron paramagnetic resonance (EPR), femtosecond transient absorption spectroscopy (FT-AS), and Kelvin probe force microscopy (KPFM) [33,34]. In the above characterization techniques, the ISIXPS and EPR spectra were widely applied to verify the carrier transmission mechanism of S-scheme heterojunction [35], which will be successively introduced by the examples below.

XPS is a progressive technique that can accurately measure the chemical shift and binding energy. Generally, for a specific semiconductor, if it receives electrons, the corresponding bind-

ing energy will shift toward higher potential; on the contrary, the binding energy will conversely shift [36]. For example, Qaraah and co-workers [37] scientifically revealed the S-scheme heterojunction charge migration mechanism between O-doped g-C₃N₄ (OCN) and N-doped Nb₂O₅ (NNBO) by an ISIXPS characterization. As presented in Fig. 5, compared with the pure NNBO, the binding energy of Nb 3d and O 1s for the OCNNb composite (without light irradiation) exhibited a shift towards lower binding energy (Fig. 5a, b), while the corresponding binding energy changes of N 1s and C 1s were opposite (Fig. 5c, d). The above changes could be mainly because when OCN and NNBO came into contact (formed heterojunction), the free electrons in OCN would transfer to the NNBO owing to their different Fermi levels, which would cause accumulations of electrons and holes onto the NNBO and OCN sides, respectively, thus generating a built-in electric field orientation from OCN to NNBO. Under light irradiation, the binding energy of Nb 3d and O 1s for the OCNNb composite exhibited a shift towards higher binding energy, whereas the corresponding changes for OCN were reversal, which primarily because under the influence of built-in electric field, the photogenerated electrons on NNBO would migrate to the OCN and occur recombination with the holes (Fig. 5e), thus retaining the carriers with powerful redox capacity to involve in interfacial catalytic reaction. Consequently, the above ISIXPS results accurately and comprehensively demonstrated that the charge migration path between NNBO and OCN obeyed S-scheme heterojunction rather than Type-II heterojunction.

Additionally, EPR spectra have also been broadly applied to reveal the carrier transmission mechanism of $g-C_3N_4$ -based S-scheme heterojunctions. For example, Fu *et al.* [22] properly



Figure 5 High-resolution XPS spectra of (a) Nb 3d, (b) O 1s, (c) N 1s, and (d) C 1s. (e) Mechanism inference and analysis. Reprinted with permission from Ref. [37]. Copyright 2022, Elsevier.

revealed that the carrier transfer path between WO₃ and g-C₃N₄ followed S-scheme mechanism *via* EPR spectra, as exhibited in Fig. 6. Evidently, the DMPO-·OH signals were sensitively captured in pure WO₃ and WO₃/g-C₃N₄ composites, while that could not be observed for pure g-C₃N₄ (Fig. 6a), mainly indicating that photogenerated holes tended to stay on the VB of WO₃ and did not migrate to the VB of g-C₃N₄. Simultaneously, the DMPO-·O₂⁻ signals could be clearly observed in pure g-C₃N₄ and WO₃/g-C₃N₄ composites, whereas a much weaker signal could be detected from pure WO₃ (Fig. 6b), suggesting that photoinduced electrons on g-C₃N₄ possessed enough reducing capacity to reduce O₂ to ·O₂⁻ and did not transmit to the CB of WO₃. As a consequence, the photoinduced carriers with pow-

erful redox capacity tended to remain on the VB of WO₃ and CB of g-C₃N₄, respectively. In other words, the carrier transmission path between WO₃ and g-C₃N₄ followed the S-scheme heterojunction mechanism (Fig. 6c) rather than Type-II heterojunctions.

In addition to the above ISIXPS and EPR characterization means, KPFM, DFT calculations, and FT-AS spectrum can also afford strong data to investigate the charge migration mechanism of g-C₃N₄-based S-scheme heterojunctions [38]. For example, KPFM can receive the variations of heterojunction surface potential with and without illumination, thus judging the carrier migration mode; FT-AS can detect the differences in absorption spectra between single components and complexes to deduce the



Figure 6 EPR signals of (a) DMPO--OH and (b) DMPO--O₂⁻. (c) Charge migration mechanism. Reprinted with permission from Ref. [22]. Copyright 2019, Elsevier.

carrier migration mechanism. In general, the above-advanced characterizations can provide sufficient and powerful evidence for the S-scheme heterojunction carrier transmission mechanism. Furthermore, more advanced characterizations need to be exploited to demonstrate the S-scheme heterojunction mechanism.

APPLICATIONS

Nowadays, $g-C_3N_4$ -based S-scheme heterojunction photocatalytic systems have been abundantly constructed and used in numerous fields because of their effective charge transmission and strong redox capability [39]. The previous reports on g- C_3N_4 -based S-scheme heterojunctions are mainly concentrated in the field of energy and environment involving H₂ production, CO₂ reduction, H₂O₂ production, and pollutant degradation [30].

H₂ evolution

Hydrogen (H₂) energy is a secondary clean energy, with high combustion value, green and clean, zero emissions, and other advantages, known as the "ultimate energy in the 21st century", but also in the context of carbon peak and carbon neutral, need to accelerate the exploitation of clean energy. Among many new technologies, photocatalytic H₂ generation refers that photocatalysts can split water into H₂ under the drive of solar energy [40]. g-C₃N₄ has been a star material in the field of hydrogen production in the past decade due to its prominent photoelectric property and suitable band structure [41]. However, the severe recombination and limited reduction ability of carriers for single-component g- C_3N_4 lead to its weak H_2 production performance. Among a variety of modification methods, the construction of g- C_3N_4 -based S-scheme heterojunctions can prominently reinforce the separation efficiency and redox capacity of carriers, thus significantly enhancing their photocatalytic activity.

Due to the relatively negative reduction potential, g-C₃N₄ is usually coupled with some oxidizing semiconductors to construct S-scheme heterojunctions, such as metal oxides, oxysalts, and organic polymers [30]. For example, Li et al. [42] fabricated a C/O doped g-C₃N₄ (COCN)/W₁₈O₄₉ heterojunction by an in-situ solvothermal method. During the solvothermal process, the nanowire-like W₁₈O₄₉ would form and *in-situ* generate onto the COCN nanosheet surface (Fig. 7a). Transmission electron microscope (TEM) images of COCN/W₁₈O₄₉ heterojunction clearly presented a typically translucent and flaky structure with several nanowire-like structures lying flat on its surfaces (Fig. 7b, c), visually proving the formation of COCN nanosheet/ W18O49 nanowire heterojunction. The as-prepared COCN/ W18O49 heterojunction displayed a prominent H2 production rate of ca. 3908.2 $\mu mol \; h^{-1} \; g^{-1}$ under simulated sunlight, which is nearly 12 times higher than that of conventional g-C₃N₄ (Fig. 7d). To reveal the mechanism of performance enhancement, the EPR spectra were carried out in Fig. 7e, f. The DMPO-·O2⁻ signals were obviously detected in COCN and COCN/



Figure 7 (a) Synthetic process and (b, c) TEM images of COCN- $W_{18}O_{49}$ heterojunction. (d) H_2 evolution rates, (e, f) EPR spectra, and (g) photocatalytic mechanism in COCN- $W_{18}O_{49}$ heterojunction. Reprinted with permission from Ref. [42]. Copyright 2023, Elsevier.

 $W_{18}O_{49}$ samples, while that could not be detected from $W_{18}O_{49}$ (Fig. 7e), powerfully indicating that photoinduced electrons of COCN possessed enough reducing capacity to reduce O_2 to $\cdot O_2^-$ and did not transfer to the CB of $W_{18}O_{49}$. However, the DMPO- \cdot OH signals were reversed (Fig. 7f), owing to that photoinduced holes tended to stay on the VB of $W_{18}O_{49}$. Evidently, the EPR results directly demonstrated that the carrier transmission path obeyed the S-scheme mechanism (Fig. 7g), which cooperated with the SPR effect to synergistically promote the enhancement of H₂-production performance.

In addition to the above combinations with oxidizing photocatalysts, $g-C_3N_4$ occasionally served as an oxidized semiconductor to design S-scheme heterojunctions with some metal sulfides. For example, Shi *et al.* [43] constructed a cobalt phthalocyanine/oxygen-doped $g-C_3N_4$ (CoPc/OCN) heterojunction photocatalyst *via* an ultrasonic method. It was found that CoPc particles were dispersedly loaded on the surface of $g-C_3N_4$ nanosheets (Fig. 8a). Moreover, the heterojunction interface between CoPc and OCN could be clearly observed from their HRTEM image in Fig. 8b. Additionally, the optimized H₂evolution rate of the resultant CoPc/OCN heterojunction reached 9560 µmol h⁻¹ g⁻¹, over 2.6-fold superior to the pure $g-C_3N_4$ (Fig. 8c). To investigate the charge transfer route of CoPc/OCN heterojunction, the KPFM potential images and their corresponding surface potential curves were supplied in Fig. 8d–f. In the dark, the surface potential of CoPc was larger than that of OCN. However, with light irradiation, the surface potential of CoPc emerged an obvious decrease, while that of OCN increased, indicating that the photoinduced electrons tended to stay on the CoPc, while the photoinduced holes stayed on the OCN. Apparently, the above KPFM results strongly suggested that the carrier transfer way between CoPc and OCN followed S-scheme heterojunction (Fig. 8g). Therefore, the greatly enhanced activity of CoPc/OCN principally profited from their generated S-scheme heterojunction.

According to the above reports and S-scheme heterojunction mechanism, it is not difficult to conclude that the interfacial electric field is the key factor driving the transmission of photoexcitation charges in S-scheme heterojunctions. Therefore, it is extremely indispensable to investigate the relationship between heterojunction electric field intensity and its corresponding photocatalytic activity. For this purpose, Zhu *et al.* [44] adopted nonmetal doping to regulate the interfacial electric field intensity of $g-C_3N_4/SnS_2$ and $g-C_3N_4/ZrS_2$ S-scheme heterojunctions and investigated the structure-activity relationship between the electric field intensity and photocatalytic activity. Specifically, compared with pristine $g-C_3N_4$, when oxygen atoms were doped into $g-C_3N_4$, the work function of resultant O-C₃N₄ would



Figure 8 (a, b) TEM images of CoPc/OCN. (c) H₂ evolution rates. (d, e) KPEM images and (f) the corresponding surface potential curves. (g) Photocatalytic charge migration mechanism. Reprinted with permission from Ref. [43]. Copyright 2023, Springer.

decrease (Fermi energy level would shift up) (Fig. 9a), which would increase the Fermi energy level difference between C₃N₄ and SnS₂, resulting in an obviously increased in the number of transferred charges (Fig. 9a), thus increasing the intensity of the interface electric field (Fig. 9b, c). Moreover, the electric field strengths of g-C₃N₄ doped with other elements (P, S) were also calculated when constructing heterojunction with SnS₂ and ZrS₂, as exhibited in Fig. 9d. Apparently, the electric field intensities of oxygen-doped g-C₃N₄ when forming heterojunction with other semiconductors were much higher than that of pristine g-C₃N₄, P-doped g-C₃N₄, and S-doped g-C₃N₄. Finally, experimental validation results indicated that the H2-evolution rate of O- C_3N_4/SnS_2 heterojunction (154 µmol h⁻¹ g⁻¹) was significantly $g-C_3N_4/SnS_2$ higher than that of heterojunction $(38 \ \mu mol \ h^{-1} \ g^{-1})$, which was consistent with the calculated results of electric field strength in Fig. 9d. Consequently, enhancing the interfacial electric field intensity of S-scheme heterojunction is an effective strategy to further enhance its photocatalytic activity, which provides a new direction for the subsequent construction of S-scheme heterojunctions.

Additionally, the most recently reported S-scheme $g-C_3N_4$ based heterojunction systems for H_2 production have been dis-

played in Table 1 [22,42-84]. On the whole, all the constructed g-C₃N₄-based S-scheme heterojunctions presented highly improved photocatalytic H₂ production performance, even some of the works have achieved an order of magnitude improvement for hydrogen production. For example, Hafeez et al. [45] prepared a rGO/NiFe₂O₄-g-C₃N₄ S-scheme heterojunction to achieve efficient hydrogen production. The as-prepared rGO/ NiFe₂O₄-g-C₃N₄ heterojunction showed an extremely prominent photocatalytic hydrogen-production rate of ca. 11,817 μ mol h⁻¹ g⁻¹, over 70 times higher than that of the g-C₃N₄ nanosheets, which was mainly owing to the fact that the formation of S-scheme heterojunction between NiFe2O4 and g-C₃N₄ could significantly accelerate charge transmission and reinforce its redox power. Moreover, the rGO/NiFe₂O₄-g-C₃N₄ was magnetic, which could be easily recovered and reused. Hassan et al. [46] fabricated a novel V₂O₅/N-deficient g-C₃N₄ Sscheme heterojunction photocatalyst by ultrasonic treatment and high-temperature calcination. The as-fabricated V₂O₅/Ndeficient $g-C_3N_4$ exhibited a prominent H_2 production rate of ca. 5892 μ mol h⁻¹ g⁻¹, which was over 13-fold superior to the g-C₃N₄. The enhanced performance could be principally attributed to the fact that the fabricated S-scheme heterojunction



Figure 9 (a) Electron migration under different Fermi levels. Photocatalytic mechanisms of (b) $g-C_3N_4/SnS_2$ and (c) $O-C_3N_4/SnS_2$ heterojunction. (d) Strength of interfacial electric field (IEF) in $g-C_3N_4/MS_2$ heterojunctions. Reprinted with permission from Ref. [44]. Copyright 2021, Elsevier.

could reinforce the carrier transmission and light absorption.

CO₂ reduction

In order to cope with the great threat to the living environment posed by rapid global climate change, developing effective CO_2 removal technologies to achieve a balanced CO_2 content in the atmosphere is desperately needed. Of all the new CO_2 removal technologies, photocatalytic CO_2 reduction can use the solar drive to reduce CO_2 into CO, CH_3OH , CH_4 , and other valuable fuels, which can not only effectively remove the CO_2 but also reduce CO_2 into valuable fuels, concurrently solving the energy and environmental crises [85]. Compared with photocatalytic H_2 production, photocatalytic CO_2 reduction requires a photocatalyst with a stronger reducing capacity. In this case, $g-C_3N_4$ has been widely investigated for photocatalytic CO_2 reduction due to its strong reducing ability, visible-light reaction, and prominent photochemical property [30]. Usually, single-constituent $g-C_3N_4$ always suffers from serious recombination of photogenerated carriers, restricted light absorption, and redox capacity, causing its weak CO_2 reduction activity. In this case, a large number of researchers solve the above defects of single g- C_3N_4 by constructing S-scheme heterojunction, so as to effectively reinforce the photocatalytic CO_2 reduction performance of g- C_3N_4 .

For instance, Wang *et al.* [86] successfully constructed a TiO₂/ g-C₃N₄ nanowire array heterojunction *via* an interesting vapour deposition method. During the vapour deposition process, the generated g-C₃N₄ thin film would *in-situ* produce onto the surface of TiO₂ nanowire array (Fig. 10a). The as-constructed TiO₂/g-C₃N₄ heterojunction displayed an excellent CO₂ reduction rate of ca. 785.3 µmol h⁻¹ g⁻¹, about 5.9 times superior to the g-C₃N₄ photocatalyst (Fig. 10b). To reveal the above performance enhancement mechanism, ISIXPS spectra were performed to demonstrate the carrier transmission route between TiO₂ and g-C₃N₄. As exhibited in Fig. 10c-f, compared to the pure g-C₃N₄, the binding energy of C 1s and N 1s for TiO₂/

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Table 1 Recently reported $g-C_3N_4$ -based S-scheme heterojunctions for H_2 production

S-scheme heterojunction	Cocatalyst (wt%)	Light source (wavelength/nm)	$\begin{array}{c} H_2 \ production \ rate \\ (\mu mol \ h^{-1} \ g^{-1}) \end{array}$	Enhancement factor <i>versus</i> g-C ₃ N ₄	Apparent quantum yield (%)	Ref.
$WO_3/g-C_3N_4$	Pt (2 wt%)	350 W Xe lamp	982	1.7	-	[22]
$g-C_3N_4/W_{18}O_{49}$	Pt (x wt%)	Simulated solar light	3908.2	11.9	13.3	[42]
CoPc/OCN	Pt (1 wt%)	300 W Xe lamp	9560	2.7	6.9	[43]
O-C ₃ N ₄ /SnS ₂	Pt (x wt%)	350 W Xe lamp ($\lambda \ge 420 \text{ nm}$)	154	4.1	_	[44]
rGO/NiFe2O4-g-C3N4	_	Solar light	11,817	76	_	[45]
V_2O_5/g - C_3N_4	Pt (5 wt%)	300 W Xe lamp ($\lambda > 420$ nm)	5892	13.1	6.5	[46]
CuInS ₂ /g-C ₃ N ₄	_	350 W Xe lamp ($\lambda > 350$ nm)	102.4	48.1	_	[47]
Ce-TiO ₂ /GO/g-C ₃ N ₄	Pt (x wt%)	300 W Xe lamp	3050	38.7	_	[48]
MnCo ₂ S ₄ /g-C ₃ N ₄	_	300 W Xe lamp	2979	26.4	0.8	[49]
WO ₃ /g-C ₃ N ₄	Pt (2 wt%)	300 W Xe lamp	2971	-	13.1	[50]
$Bi_xY_{1-x}VO_4/g-C_3N_4$	Pt (1 wt%)	Xe lamp	_	-	_	[51]
In _{2.77} S ₄ /NiS ₂ /g-C ₃ N ₄	_	300 W Xe lamp ($\lambda > 390$ nm)	7481.7	52.5	0.3	[52]
NiTiO ₃ /g-C ₃ N ₄	Pt (x wt%)	2×250 W Tungsten-halogen lamp	576	1.4	_	[53]
AgI/g-C ₃ N ₄	Pt (0.6 wt%)	Xe lamp ($\lambda \ge 420 \text{ nm}$)	4562	10.6	3.2	[54]
In ₂ S ₃ /g-C ₃ N ₄ /CoZnAl- LDH	_	300 W Xe lamp	50.6	-	_	[55]
Co_3O_4/g - C_3N_4	_	300 W Xe lamp	105.1	41	0.05	[56]
Co-Sn ₃ O ₄ /g-C ₃ N ₄	Pt (3 wt%)	300 W Xe lamp ($\lambda > 420$ nm)	1793.9	1.6	_	[57]
$TiO_2/g-C_3N_4$	Pt (1 wt%)	$4 \times \text{LED}^{a} \text{ lamp } (\lambda = 420 \text{ nm})$	3211	7.5	1.6	[58]
$g-C_3N_4/BiO_{1.2}I_{0.6}$	_	500 W Xe lamp ($\lambda > 420$ nm)	1402.7	3.5	11.8	[59]
$S\text{-}g\text{-}C_3N_4/g\text{-}C_3N_4$	Pt (3 wt%)	300 W Xe lamp ($\lambda \ge 420 \text{ nm}$)	5548.1	60.8	0.4	[60]
$Mn_{0.2}Cd_{0.8}S$ -D/g-C ₃ N ₄	Pt (x wt%)	300 W Xe lamp ($\lambda > 420$ nm)	11,420	30	_	[61]
Bi ₃ TaO ₇ /g-C ₃ N ₄	Pt (1 wt%)	300 W Xe lamp ($\lambda > 420$ nm)	4891	3	4.1	[62]
$Ni_{0.85}Se/g-C_3N_4$	_	300 W Xe lamp	8780.3	92.9	_	[63]
MnWO ₄ /g-C ₃ N ₄	Pt (0.5 wt%)	200 W Xe lamp	871.4	3.7	16.5	[64]
NiTe ₂ /g-C ₃ N ₄	Pt (1 wt%)	300 W Xe lamp	2540.4	23.4	_	[65]
TiO_2 -OV/g-C ₃ N ₄	Pt (1.2 wt%)	300 W Xe lamp ($\lambda > 400 \text{ nm}$)	1096	10.9	_	[66]
g-C ₃ N ₄ /CdSe-D	Pt (x wt%)	300 W Xe lamp ($\lambda > 420$ nm)	18,800	1446	38.4	[67]
$Cu_2O/g-C_3N_4$	_	500 W Xe lamp ($\lambda > 400$ nm)	480.6	4.6	_	[69]
g-C ₃ N ₄ /CdS	Pt (3 wt%)	300 W Xe lamp	3370	3.8	_	[70]
SbVO ₄ /g-C ₃ N ₄	-	300 W Xe lamp	752	4.1	_	[71]
Ni - Sn_3O_4/g - C_3N_4	Pt (3 wt%)	300 W Xe lamp ($\lambda > 420$ nm)	1961	1.4	_	[72]
$Mn_{0.5}Cd_{0.5}Se/g-C_3N_4$	-	280 W Xe lamp	5908.3	126.6	_	[73]
Ni ₂ P/g-C ₃ N ₄ /Cd _{0.5} Zn _{0.5} Se-D	Pt (1 wt%)	300 W Xe lamp ($\lambda \ge 420$ nm)	12,627	-	37.7	[74]
N-MoS ₂ /S-g-C ₃ N ₄	-	300 W Xe lamp	658.5	23	_	[75]
$S-g-C_3N_4/WO_{2.72}$	_	300 W Xe lamp ($\lambda > 420$ nm)	786	-	7.6	[76]
CdS/g-C ₃ N ₄	-	300 W Xe lamp	15,300	3060	6.9	[77]
CdS/g-C ₃ N ₄ -GA	-	300 W Xe lamp ($\lambda > 420$ nm)	86.4	3.5	_	[78]
g-C ₃ N ₄ /CdS-DETA	Pt (0.6 wt%)	300 W Xe lamp ($\lambda > 400 \text{ nm}$)	9738	12.2	10.2	[79]
MCN/UCN	Pt (1 wt%)	300 W Xe lamp ($\lambda > 420$ nm)	598	-	1.1	[80]
$W_{18}O_{49}/g$ - C_3N_4	Pt (3 wt%)	300 W Xe lamp ($\lambda > 420$ nm)	4670	15.1	9.8	[81]
ZnCdS/DBTCN	Pt (1 wt%)	300 W Xe lamp	8870	6.6	14.9	[82]
$NiCo_2O_4/g-C_3N_4$	Pt (0.5 wt%)	300 W Xe lamp	424	4.2	-	[83]
Bi ₂ MoO ₆ SOVs/g-C ₃ N ₄	_	300 W Xe lamp ($\lambda \ge 420 \text{ nm}$)	2290	12.1	_	[84]

a) LED: light-emitting diode.

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Figure 10 (a) Structure diagram of $TiO_2/g-C_3N_4$ heterojunction. (b) CO_2 reduction activity. (c-f) *In-situ* XPS spectra and (g) graphic illustration for the S-scheme mechanism of $TiO_2/g-C_3N_4$. Reprinted with permission from Ref. [86]. Copyright 2022, Elsevier.

g-C₃N₄ heterojunction (without light irradiation) exhibited a shift towards higher binding energy (Fig. 10c, d), while the corresponding binding energy changes of Ti 2p and O 1s were opposite (Fig. 10e, f). The above shifts mainly because when TiO₂ and g-C₃N₄ came into contact (formed heterojunction), the free electrons in g-C₃N₄ would transfer to the TiO₂ because of their different work function, which would cause accumulations of electrons and holes onto the TiO₂ and g-C₃N₄ sides, respectively, thus generating a built-in electric field orientation from g-C₃N₄ to TiO₂ (Fig. 10g). Under light irradiation, the binding energy of C 1s and N 1s for the TiO₂/g-C₃N₄ composite exhib-

ited a shift towards higher binding energy, whereas the corresponding changes for TiO₂ were reversal, primarily because that with the action of a built-in electric field, the photogenerated electrons on TiO₂ would migrate to the g-C₃N₄ and occur recombination with the holes (Fig. 10g), thus retaining the carriers with powerful redox capacity to involve in interfacial catalytic reaction. Therefore, the generated TiO₂/g-C₃N₄ S-scheme heterojunction greatly contributed to their reinforced catalytic activity.

Apart from TiO_2 , $g-C_3N_4$ is also formed S-scheme heterojunctions with other oxides. For example, Dai *et al.* [87] pre-

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pared a Cu plasmonic-modified $g-C_3N_4/Cu_2O$ S-scheme heterojunction photocatalyst by a two-step procedure, involving the initial wet chemistry process and the following in-situ chemical reduction. The $g-C_3N_4$, Cu_2O , and Cu could be clearly identified from the TEM and high resolution TEM (HRTEM) images of $g-C_3N_4/Cu_2O@Cu$ composite (Fig. 11a-c). Moreover, the asobtained $g-C_3N_4/Cu_2O@Cu$ sample emerged with a prominent CO_2 reduction rate of about 18.9 µmol h^{-1} g^{-1} , which was obviously higher than that of pure $g-C_3N_4$ and Cu_2O (Fig. 11d), respectively. To disclose the charge transfer way, the EPR spectra were conducted, and the results were presented in Fig. 11e. Evidently, the DMPO- \cdotO_2^- signals could be easily tested for the pure $g-C_3N_4$ and $g-C_3N_4/Cu_2O@Cu$ composites, whereas

a much weaker signal could be detected from pure Cu₂O, indicating that photoinduced electrons on $g-C_3N_4$ possessed enough reducing capacity and did not transport to the CB of Cu₂O. Namely, the charge transfer path between Cu₂O and $g-C_3N_4$ followed S-scheme heterojunction mechanism (Fig. 11f) rather than Type-II heterojunctions. Consequently, the above significantly enhanced CO₂ reduction activity could be mainly due to the synergistic promotion effects of the plasma resonance effect of Cu and the formed $g-C_3N_4/Cu_2O$ S-scheme heterojunction (Fig. 11f).

In addition to the above combinations with oxidizing photocatalysts, $g-C_3N_4$ occasionally served as an oxidized semiconductor to construct S-scheme heterojunctions with some



Figure 11 (a) TEM and (b, c) HRTEM images of $g-C_3N_4/Cu_2O@Cu$. (d) CO_2 reduction activity. (e) EPR spectra and (f) photocatalytic mechanism. Reprinted with permission from Ref. [87]. Copyright 2022, Elsevier.

metal sulfides. For instance, Lee and co-workers [88] designed a S-doped Cu₃P/g-C₃N₄ S-scheme heterojunction through a facile liquid-phase mixing method. Apparently, the Cu₃P emerged as an ultra-small nanoparticle structure with a size of less than 10 nm and attached to g-C₃N₄ surface (Fig. 12a). Moreover, the as-prepared Cu₃P/g-C₃N₄ composite showed an evidently enhanced CO₂ reduction rate of around 137 μ mol h⁻¹ g⁻¹, which is nine times superior to the pure $g-C_3N_4$ (Fig. 12b). To analyze the above performance enhancement mechanism, the EPR spectra were carried out and presented in Fig. 12c. In contrast to the pure g-C₃N₄, the Cu₃P and Cu₃P/g-C₃N₄ samples exhibited much stronger DMPO- \cdot O₂⁻ signals, revealing that photoinduced electrons on Cu₃P possessed enough reducing capacity to involve in reduction reaction and did not migrate to the CB of g-C₃N₄. In other words, the carrier transmission way between Cu₃P and g-C₃N₄ followed the S-scheme heterojunction mechanism (Fig. 12d), contributing to their significantly improved CO₂ reduction performance.

In addition, the most recently reported g-C₃N₄-based S-scheme heterojunctions for CO₂ reduction have been displayed in Table 2 [32,37,86–100]. Generally, g-C₃N₄ could be fabricated S-scheme heterojunctions with many other semiconductors due to its excellent band structure. For instance, Bashal *et al.* [89] synthesized a Cu nanoparticle-modified g-C₃N₄/MoS₂ S-scheme heterojunction photocatalyst. The asprepared Cu/g-C₃N₄/MoS₂ composite presented an extremely strong CO₂ reduction rate of ca. 146.7 µmol h⁻¹ g⁻¹, over 9-fold higher than that of g-C₃N₄. Tahir *et al.* [94] developed a Ti₃AlC₂ MAX cocatalyst-modified g-C₃N₄/TiO₂ heterojunction photocatalyst *via* a sol-gel dip-coating strategy. The resultant Ti₃AlC₂ MAX-g-C₃N₄/TiO₂ composite exhibited a significantly rein-

forced CO₂ reduction activity of ca. 2400.7 μ mol h⁻¹ g⁻¹, which is over 23 times superior to the pure g-C₃N₄. The above evidently reinforced CO₂ reduction performance could be mainly due to the fact that the charge transfer route of the generated g-C₃N₄/ TiO₂ heterojunction obeyed the S-scheme mechanism.

Pollutant degradation

With the advancement of global industrialization, environmental pollution has become increasingly serious, and the content of various pollutants, such as oxynitrides, antibiotics, and dyes, in the atmosphere, water, and soil has seriously exceeded the standard [101]. Therefore, it is urgent to seek effective methods to remove these environmental pollutants. Among the many processing strategies, photocatalytic degradation technique can effectively remove the pollutants in the atmosphere, water, and soil, and the degradation process of pollutants is mild and does not produce toxic substances. Moreover, the effect of the photocatalyst cycle is not reduced, and it can be recycled and reused. Based on the photocatalytic degradation mechanism, the more positive the VB of the semiconductor, the more stronger oxidation capacity of its photogenerated holes, thus efficiently degrading various pollutants into non-toxic and harmless CO2 and H2O. However, g-C3N4, a typical RP, usually presents very weak photocatalytic degradation activity due to its limited oxidation capacity and severe recombination of carriers. In this case, combining the OPs with g-C₃N₄ to form S-scheme heterojunctions could effectively solve the problem of weak oxidation capacity and severe recombination of carriers for g-C₃N₄, thus greatly enhancing its photocatalytic degradation activity.

For instance, Van Pham et al. [102] constructed a g-C₃N₄/



Figure 12 (a) TEM and HRTEM (inset) images of us- $Cu_3P/S/CN$. (b) CO_2 reduction activity. (c) EPR spectra and (d) S-scheme mechanism. Reprinted with permission from Ref. [88]. Copyright 2021, American Chemical Society.

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Table 2 Recently reported g-C₃N₄-based S-scheme heterojunctions for CO₂ reduction

S-scheme heterojunction	Light source (wavelength/nm)	Reduction products	CO_2 reduction rate (µmol h ⁻¹ g ⁻¹)	Enhancement factor <i>versus</i> g-C ₃ N ₄	Apparent quantum yield (%)	Ref.
InVO ₄ QDs/g-C ₃ N ₄	300 W Xe lamp (λ > 420 nm)	CO (93.3%)	69.8	18	6.8	[32]
O-g-C ₃ N ₄ /N-Nb ₂ O ₅	300 W Xe lamp	CO (78.82%) CH ₄ (21.18%)	321.5	6	4.9	[37]
$TiO_2/g-C_3N_4$	300 W Xe lamp	CO CH ₄	785.3	5.9	_	[86]
g-C ₃ N ₄ /Cu ₂ O@Cu	300 W Xe lamp	CO CH ₄	13.9	-	_	[87]
$Cu_3P/g-C_3N_4$	300 W Xe lamp	СО	137	9	_	[88]
g-C ₃ N ₄ /MoS ₂ /Cu	350 W Xe lamp (λ > 420 nm)	CO (100%)	146.7	9.7	3.2	[89]
g-C ₃ N ₄ /BiOBr	300 W Xe	CH ₃ OH	267	_	_	[90]
g-C ₃ N ₄ /Pd/MoO _{3-x}	300 W Xe lamp	СО	4.6	12.1	0.01	[91]
$Ti_3C_2\text{-}g\text{-}C_3N_4/MoSe_2$	Simulated sunlight	CO CH ₄	47.8	-	_	[92]
g-C ₃ N ₄ /Ag-TiO ₂	300 W Xe lamp	CO CH ₄	52.7	-	2.4	[93]
g-C ₃ N ₄ /TiO ₂ /Ti ₃ AlC ₂ MAX	35 W HID car lamp	CO (3.41%) CH ₄ (96.59%)	2400.8	23.3	_	[94]
$ZnIn_2S_4/g$ - C_3N_4	300 W Xe lamp	СО	883	13.4	8.9	[95]
CsPbBr ₃ /S-g-C ₃ N ₄	300 W Xe lamp (λ > 400 nm)	СО	83.6	4	_	[96]
Bi ₃ NbO ₇ /g-C ₃ N ₄	Simulated solar	CH ₄ (90%)	37.6	15	_	[97]
BiOI/g-C ₃ N ₄	300 W Xe lamp (λ > 400 nm)	СО	3.1	5.4	_	[98]
g-C ₃ N ₄ /ZnO	300 W Xe lamp	CH_4	16	7	_	[99]
rGO/R-CeO ₂ /g-C ₃ N ₄	300 W Xe lamp	CO CH ₄	23.9	4.6	_	[100]

SnO₂ S-scheme heterojunction *via* a facile calcination process, involving the pre-synthesizing SnO₂ nanoparticles and g-C₃N₄ nanosheets by simple hydrothermal and calcination treatments, respectively (Fig. 13a). The HRTEM image of g-C₃N₄/SnO₂ composite clearly emerged that SnO₂ nanoparticles with different sizes were loaded on g-C₃N₄ nanosheet surface (Fig. 13b), investigating the successful production of g-C₃N₄/SnO₂ heterojunction. Additionally, the as-constructed g-C₃N₄/SnO₂ heterojunction showed obviously reinforced photocatalytic NO removal activity, which is 1.2 times higher than that of pure g-C₃N₄ (Fig. 13c). The above enhanced photocatalytic activity could be primarily owing to the fact that the generated g-C₃N₄/ SnO₂ S-scheme heterojunction could effectively promote carrier transmission and enhance its oxidation capacity (Fig. 13d).

Generally, semiconductors with strong oxidation ability are mostly metal oxides. Hence, $g-C_3N_4$ is usually combined with various metal oxides to construct S-scheme heterojunctions and applied in photocatalytic degradation. For another example, Truong and co-workers [103] fabricated a novel α -Fe₂O₃/g-C₃N₄ S-scheme heterojunction by a simple sonication treatment, including the pre-synthesizing α -Fe₂O₃ and g-C₃N₄ via sol-gel method and calcination process (Fig. 14a). The as-fabricated α -Fe₂O₃/g-C₃N₄ heterojunction displayed prominently reinforced photocatalytic degradation performance of cefalexin and amoxicillin, which was 5-fold and 9-fold superior to the pure $g-C_3N_4$, respectively (Fig. 14b, c). To reveal the above performance enhancement mechanism, EPR spectra were conducted. Obviously, under visible light, the DMPO- $\cdot O_2^-$ and DMPO- $\cdot OH$ signals could be clearly detected in $g-C_3N_4$ and α -Fe₂O₃/ $g-C_3N_4$ samples, while they could not be detected in dark (Fig. 14d, e). Combining the active-radical trapping test results, it was not difficult to draw a conclusion that the carrier transmission way obeyed the S-scheme heterojunction (Fig. 14f), which contributed to the significantly enhanced degradation rate of α -Fe₂O₃/ $g-C_3N_4$.

Except for metal oxides, some oxysalt semiconductors are also often used to construct S-scheme heterojunctions with g- C_3N_4 . For instance, Dai *et al.* [104] produced an Au/g- C_3N_4 /BiO_{1,2}I_{0.6} photocatalyst (Fig. 15a) by a facile calcination method. The TEM and HRTEM images of the resultant Au/g- C_3N_4 /BiO_{1,2}I_{0.6} sample clearly showed the lattice fringes of characteristic crystal faces for Au and BiO_{1,2}I_{0.6} (Fig. 15b, c). Moreover, the semitransparent and amorphous lamellar structures of g- C_3N_4 could be observed, powerfully indicating the successful production of Au/g- C_3N_4 / BiO_{1,2}I_{0.6} heterojunction structure. Additionally, the as-designed Au/g- C_3N_4 /BiO_{1,2}I_{0.6} sample displayed excellent photoreduction activity of Cr(VI) and photocatalytic degradation activity of BPAF, which were 3.7 and 6.5 times higher than that of the pure



Figure 13 (a) Synthetic process and (b) HRTEM image of $g-C_3N_4/SnO_2$ somposite. (c) NO degradation rate constants and (d) photocatalytic reaction mechanism. Reprinted with permission from Ref. [102]. Copyright 2021, Elsevier.

g-C₃N₄ (Fig. 15d, e), respectively. The characterization and theoretical calculation results indicated that the plasmon resonance effect of Au and the formation of S-scheme heterojunction jointly promoted the enhancement of photocatalytic performance for Au/g-C₃N₄/BiO_{1.2}I_{0.6} sample.

In addition to the above examples, the most recently reported $g-C_3N_4$ -based S-scheme heterojunctions for photocatalytic degradation contaminant are shown in Table 3 [68,102–163]. It is found that photocatalytic degradation technology can be applied to degrade various antibiotics, dyes, atmospheric pollutants, and even remove toxic heavy metal ions. Moreover, most of the reported $g-C_3N_4$ -based S-scheme heterojunctions show extremely increased photocatalytic degradation activity. For instance, Mkhalid *et al.* [105] prepared a $g-C_3N_4/Li_2MnO_3$

heterojunction photocatalyst by a simple evaporation process. The obtained g-C₃N₄/Li₂MnO₃ heterojunction exhibited an obviously reinforced photocatalytic degradation rate of trichloroethylene, which was over 5-fold superior to the pure g-C₃N₄. Moreover, the improvement was mainly owing to the generation of S-scheme heterojunction, which significantly promoted the separation and transmission of photoinduced charges. Van et al. [106] constructed a g-C₃N₄/CdS nano-heterojunction *via* a solvothermal method. The as-prepared g-C₃N₄/CdS heterojunction emerged with an extremely increased photocatalytic degradation rate of methylene blue, nearly 25 times higher than that of pure g-C₃N₄, primarily due to the construction of S-scheme heterojunction between g-C₃N₄ and CdS.



Figure 14 (a) Synthetic process of α -Fe₂O₃/g-C₃N₄. Photocatalytic degradation of (b) cefalexin and (c) amoxicillin. EPR signals of (d) DMPO- O_2^- and (e) DMPO- O_1 . (f) Photocatalytic mechanism. Reprinted with permission from Ref. [103]. Copyright 2022, American Chemical Society.



Figure 15 (a) Graphical illustration. (b) TEM and (c) HRTEM images of $Au/g-C_3N_4/BiO_{1.2}I_{0.6}$. Kinetic fitting photodegradation curves of (d) Cr(VI) and (e) BPAF. (f) Photocatalytic mechanism. Reprinted with permission from Ref. [104]. Copyright 2022, Elsevier.

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Table 3 Recently reported $g-C_3N_4$ -based S-scheme heterojunctions for pollutant degradation

S-scheme heterojunction	Light source (wavelength/nm)	Degradation substrate	Reduction rate (min ⁻¹)	Enhancement factor <i>versus</i> g-C ₃ N ₄	Ref.	
$Ti_{0.7}Sn_{0.3}O_2/g\text{-}C_3N_4$	1.5 W LED lamp	Rhodamine B Tetracycline hydrochloride	0.205 0.053	2.2 8.8	[68]	
$g-C_3N_4/SnO_2$	300 W Xe lamp (400 < λ < 800 nm)	NO	0.0881	1.2	[102]	
a-FeaOa/a-CaN	300W Osram lamp	Cefalexin	0.0113	5.0	[103]	
u-1 c ₂ O ₃ /g-O ₃ iv ₄	$(\lambda > 420 \text{ nm})$	Amoxicillin	0.004	9.0	[105]	
Au/g-C ₃ N ₄ /BiO _{1.2} I _{0.6}	500 W Xe lamp	Bisphenol AF	0.0174	6.5	[104]	
0 0 0 1 0 1 0 0 0	$(\lambda > 420 \text{ nm})$	Cr (VI)	0.0204	3.7		
$Li_2MnO_3/g-C_3N_4$	$300 \text{ W Xe lamp} \\ (\lambda > 420 \text{ nm})$	Trichloroethylene	0.0127	6.7	[105]	
CdS/g-C ₃ N ₄	30 W LED lamp	Methylene blue	0.025	25	[106]	
$Bi_2MoO_6(2 \ 0 \ 0)/g-C_3N_4$	Xe lamp $(\lambda > 420 \text{ nm})$	Hg^0	-	-	[107]	
R.palustris/RCM@CPU	500 W Xe lamp (λ > 420 nm)	Azo dye	-	-	[108]	
σ-CaNt/TiOa	300 W Xe lamp	Methylene Blue	0.0548	5.4	[109]	
g 03114/1102	500 W Ac lamp	Tetracycline	0.3154	8.5		
$TiO_2/g-C_3N_4$	350 W Xe lamp	LBW	0.0023	2.9	[110]	
$g-C_3N_4/Mn(VO_3)_2$	500 W Xe lamp	Sulfamethoxazole	-	14	[111]	
LaFeO ₃ /g-C ₃ N ₄	1000 W Xe lamp	RB-19	0.0239	24.8	[112]	
	r	Cr (VI)	0.0172	10.3		
$ZnFe_2O_4/g-C_3N_4$	300 W Xe lamp	Tetracycline hydrochloride	0.0454	4.6	[113]	
g-C ₃ N ₄ /BiOI	Xe lamp	Tetracycline	0.1687	27.2	[114]	
0.5.	$(\lambda > 420 \text{ nm})$	Cr (VI)	0.0718	8.5		
g-C ₃ N ₄ /NiFe ₂ O ₄	300 W Xe lamp	Tetracycline	0.0145	2.3	[115]	
CeO_2 -C-g- C_3N_4	250 W Xe lamp (320 < λ < 780 nm)	Methylene blue Tetracyclines	0.0567 0.0194	2.4 1.5	[116]	
TiO _{2-x} /g-C ₃ N ₄ /CNFe	300 W Xe lamp ($\lambda > 420 \text{ nm}$)	Peroxymonosulfate	0.0969	_	[117]	
g-C ₃ N ₄ /WO ₃ /ZnS	300 W Xe lamp (λ > 420 nm)	Tetracycline	0.042	3.5	[118]	
O-g-C ₃ N ₄ /OV BiOCl	500 W Xe lamp (λ > 400 nm)	Bisphenol A	0.035	12.1	[119]	
$MgO/g-C_3N_4$	300 W Xe lamp (λ > 420 nm)	Rhodamine B	0.0064	33.7	[120]	
		Tetracycline	_	-		
$g-C_3N_4/GO/ZnFe_2O_4$	Halogen lamp	Rhodamine B	-	_	[121]	
		Methylene Blue	-	-		
g-C ₃ N ₄ /CeO ₂	300 W Xe lamp	Bisphenol A	0.0257	8.6	[122]	
g-C3N4/TiO2/ZnIn2S4/GA	300 W Xe lamp	Methyl orange	0.094	37.6	[123]	
0 0 0 0 0 0 0 0 0 0	· · · · · · · · · · · · · · · · · · ·	Cr (VI)	0.0411	9.3		
$La_2Ce_2O_7/g\text{-}C_3N_4$	$300 \text{ W Xe lamp} \\ (\lambda > 420 \text{ nm})$	Rhodamine B	0.027	2.4	[124]	
g-C ₃ N ₄ /TiOF ₂	500 W Xe lamp	Tetracycline hydrochloride	1.052	2.8	[125]	
$N\text{-}TiO_{2-X}/g\text{-}C_3N_4$	300 W Xe lamp $(\lambda > 400 \text{ nm})$	2,4-dinitrophenylhydrazine	-	-	[126]	
α -Fe ₂ O ₃ /g-C ₃ N ₄	300 W Xe lamp	Tetracycline hydrochloride	0.0165	3.1	[127]	
$Ag_2CO_3/Bi_4O_5I_2/g\text{-}C_3N_4$	300 W Xe lamp	Tetracycline	0.0389	13.3	[128]	
N-g-C ₃ N ₄ /NH ₂ -MIL-125(Ti)	300 W Xe lamp (λ > 400 nm)	Rhodamine B	0.0246	3.3	[129]	
CaSnO ₃ /g-C ₃ N ₄	500 W Halogen lamp	Methylene Blue	-	-	[130]	

(To be continued on the next page)

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(Continued)

S-scheme heterojunction	Light source (wavelength/nm)	Degradation substrate	Reduction rate (min ⁻¹)	Enhancement factor <i>versus</i> g-C ₃ N ₄	Ref.
$SnO_{2-x}/g-C_3N_4$	300 W Osram lamp ($\lambda > 420 \text{ nm}$)	NO	-	-	[131]
g-C ₃ N ₄ /Ag/AgNCO	300 W Xe lamp	Tetracycline	0.3	3.1	[132]
$g-C_3N_4/SnO_2$	300 W Xe lamp	NO	-	-	[133]
$\alpha\text{-}Fe_2O_3/g\text{-}C_3N_4$	300 W Xe lamp ($\lambda > 420 \text{ nm}$)	Tetracycline	-	-	[134]
$N-ZnO/g-C_3N_4$	300 W Xe lamp	Norfloxacin	0.034	4.1	[135]
$SnO_2/g-C_3N_4$	30 W LED lamp	Rhodamine B	0.0353	10.7	[136]
$PDI/g\text{-}C_3N_4/TiO_2@Ti_3C_2$	300 W Xe lamp ($\lambda > 420 \text{ nm}$)	Atrazine	0.0248	4.4	[137]
$WO_3/g-C_3N_4$	300 W Solar simulation light	NO	0.119	2.1	[138]
g - $C_3N_4/BiVO_4$	300 W Xe lamp (λ > 400 nm)	Methylene blue	0.0609	15.2	[139]
MoO ₃ /g-C ₃ N ₄	300 W Xe lamp	Rhodamine B Cr (VI)	-	-	[140]
g-C ₃ N ₄ /TiO ₂ /CFs	350 W Xe lamp	Tetracycline hydrochloride	0.1566	9.5	[141]
DOD: NI: D/r C N	300 W Xe lamp	Rhodamine B	0.026	3.6	[142]
$B1OBT/N1_2P/g-C_3N_4$	$(\lambda \ge 400 \text{ nm})^{-1}$	Methyl orange	0.0154	3.9	[142]
Bi_2Se_3/g - C_3N_4	60 W LED lamp $(\lambda > 400 \text{ nm})$	Phenol	-	-	[143]
$g-C_3N_4/TiO_2$	UV light	Tetracycline hydrochloride	0.057	-	[144]
TiO ₂ /CN QDs	500 W Xe lamp	Methyl orange	0.0172	-	[145]
g-C ₃ N ₄ /Mo-WO ₃	350 W Xe lamp	Methyl orange	0.0367	14.1	[146]
$g\text{-}C_3N_4/ZnIn_2S_4$	LED lamp $(\lambda > 400 \text{ nm})$	Tetracycline	0.0196	2.5	[147]
g-C₂N₄/NiZnAl-LDH	500 W Xe lamp (λ < 400 nm)	Methyl orange	0.0233	3.5	[148]
8 - 5 - 14 - 1		Tetracycline	0.0145	3.2	[]
g-C ₃ N ₄ /rGO/ZnO-Ag	_	Rhodamine B	0.017	5.7	[149]
0 0 0		Methyl orange	0.023	4.9	
BiOBr/g-C ₃ N ₄	500 W Xe lamp ($\lambda > 400 \text{ nm}$)	Ethyl xanthate	0.0282	24.6	[150]
$SnS_2/g-C_3N_4$	60 W lamp $(200 < \lambda < 400 \text{ nm})$	Rhodamine B	0.0074	4.2	[151]
$CoTiO_3/g-C_3N_4$	$300 \text{ W Xe lamp} \\ (\lambda > 400 \text{ nm})$	Methyl orange	0.0165	38.1	[152]
g-C ₃ N ₄ /MXene/Ag ₃ PO ₄	$\begin{array}{l} 300 \text{ W Xe lamp} \\ (\lambda \geq 420 \text{ nm}) \end{array}$	Tetracycline	0.0353	4.3	[153]
$ZnFe_2O_4/g\text{-}C_3N_4$	$\begin{array}{l} 300 \text{ W Xe lamp} \\ (\lambda \geq 400 \text{ nm}) \end{array}$	Bisphenol A	0.075	-	[154]
$Bi_4V_2O_{11}/g\text{-}C_3N_4$	$300 \text{ W Xe lamp} \\ (\lambda > 420 \text{ nm})$	Oxytetracycline	-	3.3	[155]
BiOBr/g-C ₃ N ₄	$300 \text{ W Xe lamp} \\ (\lambda > 400 \text{ nm})$	Rhodamine B	0.0127	48.2	[156]
$ZnFe_2O_4/g-C_3N_4$	LED lamp	Uranium (VI)	-	-	[157]
Bi_2WO_6/g - C_3N_4	300 W Xe lamp ($\lambda > 400 \text{ nm}$)	Ammonium dinitramide	0.0567	44.3	[158]
$S-g-C_3N_4/TiO_2$	300 W Xe lamp	Congo Red	0.0962	8.2	[159]
$Cd_{0.5}Zn_{0.5}S/g\text{-}C_{3}N_{4}$	350 W Xe lamp ($\lambda > 420 \text{ nm}$)	Rhodamine B	0.0817	13	[160]
$C-WO_3/g-C_3N_4$	$300 \text{ W Xe lamp} \\ (\lambda > 420 \text{ nm})$	Tetracycline	0.0378	2.3	[161]
$Bi_2MoO_6/g-C_3N_4$	$300 \text{ W Xe lamp} \\ (\lambda > 420 \text{ nm})$	Rhodamine B	0.0808	9.7	[162]
$g-C_3N_4/\alpha-Fe_2O_3$	Halogen lamp	Methyl orange	0.029	-	[163]



Figure 16 (a) Preparation process. (b, c) HRTEM images. (d) H_2O_2 production rates. Work functions of (e) $g-C_3N_4$ and (f) CdS. (g) S-scheme mechanism. Reprinted with permission from Ref. [165]. Copyright 2023, American Chemical Society.

H_2O_2 production

Hydrogen peroxide (H_2O_2) , a powerful oxidizing agent and bleach, has been broadly used in industry, aerospace, energy, and other fields due to its advantages of pollution-free, green, and clean [164]. Currently, the anthraquinone method is mainly used to produce H_2O_2 . However, the anthraquinone method always causes high energy consumption, large carbon, organic solvents and pollutants emission. Therefore, developing environmentally friendly, efficient, and economical strategy to produce H_2O_2 is just what the public wanted. Based on the fact, photocatalytic H_2O_2 production can convert H_2O and O_2 into H_2O_2 driven by sunlight. Namely, H_2O_2 can be efficiently produced by a suitable photocatalyst [85]. According to the mechanism of photocatalytic production H_2O_2 , currently reported H_2O_2 generation is mainly based on photogenerated electron reduction reaction. In this case, g- C_3N_4 has been broadly used to produce H_2O_2 .

For example, Phan *et al.* [165] designed a $g-C_3N_4/CdS$ S-scheme heterojunction *via* a plain calcination strategy (Fig. 16a). The HRTEM images of $g-C_3N_4/CdS$ composite indicated that CdS particles were aggregated on the surface of amorphous $g-C_3N_4$. Moreover, the lattice fringes corresponding to (103), (101), and (200) crystal faces could be clearly observed (Fig. 16b, c). Additionally, the resultant $g-C_3N_4/CdS$ hetero-

junction emerged with a prominent H_2O_2 production rate of 23,440 µmol h⁻¹ g⁻¹, which was over two times superior to the pure g-C₃N₄ (Fig. 16d), primarily owing to the generated g-C₃N₄/CdS S-scheme heterojunction. To demonstrate the formation process of S-scheme heterojunction, the work functions of g-C₃N₄ and CdS were calculated and shown in Fig. 16e, f. Due to their different work functions, the free electrons of CdS would migrate to the g-C₃N₄ when they came into contact, thus generating a built-in electric field direction from CdS to g-C₃N₄ (Fig. 16g). When they were excited by the light, the photogenerated carrier with weak redox ability would recombine, while the carrier with strong redox ability would be retained and participated in the following interface reaction, thus contributing to their highly increased performance.

Except for metal sulfides, $g-C_3N_4$ is also often coupled with some metal oxides to construct S-scheme heterojunction and used to produce H_2O_2 . For instance, Wang and co-workers [166] designed a Pd-modified $Cu_2O/g-C_3N_4$ S-scheme heterojunction photocatalyst. The TEM image revealed that the Cu_2O -Pd particles were glomerated onto the surface of the semitransparent $g-C_3N_4$ framework (Fig. 17a). Moreover, the lattice fringes and spacings corresponding to Cu_2O and Pd could be clearly observed from their HRTEM images in Fig. 17b, c. Simulta-



Figure 17 (a) TEM and (b, c) HRTEM images of $g-C_3N_4/Cu_2O-Pd$ sample. (d) H_2O_2 production rates and (e) H_2O_2 production and charge transfer mechanism. Reprinted with permission from Ref. [166]. Copyright 2023, American Chemical Society.

neously, the lattice fringes of Cu₂O and Pd emerged obvious intersections (Fig. 17c), adequately indicating the successful generation of Pd-modified Cu₂O/g-C₃N₄ heterojunction structures. Additionally, the resultant g-C₃N₄/Cu₂O-Pd sample exhibited a super-strong H₂O₂ production rate of 34,000 µmol h⁻¹ g⁻¹, which was over 10-fold stronger than that of pure g-C₃N₄ (Fig. 17d). The greatly improved performance can be mainly ascribed to the synergistic promotion of S-scheme heterojunction and Pd modification (Fig. 17e).

In addition to the above metal oxides and sulfides, metalorganic frameworks have been broadly combined with g-C₃N₄ to produce heterojunction photocatalysts. For example, Xia et al. [167] fabricated a novel g-C₃N₄/Zinc porphyrin (Zn-TCPP) Sscheme heterojunction photocatalyst by a thermal polycondensation route. The as-prepared g-C₃N₄/Zn-TCPP heterojunction composite presented an evidently improved H2O2 production-rate of 355.13 µmol h⁻¹ g⁻¹, over 3-fold superior to the g-C₃N₄ (Fig. 18a). Moreover, the g-C₃N₄/Zn-TCPP could maintain its initial high performance after four H₂O₂-production cycles (Fig. 18b). To demonstrate the charge transfer route between g-C₃N₄ and Zn-TCPP, KPFM technology was conducted, and its corresponding results were provided in Fig. 18c-f. In dark, the surface potential of Zn-TCPP (point B, 402 mV) was obviously higher than that of $g-C_3N_4$ (point A, 101 mV), which would cause the migration of electrons from Zn-TCPP to $g-C_3N_4$, thus producing a built-in electric field of direction from Zn-TCPP to g-C₃N₄ (Fig. 18g). With light illumination, the surface potential of g-C₃N₄ (point A) increased even more, due to the fact that the photoinduced electrons migrated from g-C₃N₄ to Zn-TCPP, consistent with the direction of the electric field. Consequently, the above KPFM results *in-situ* confirmed the carrier transmission path of S-scheme heterojunction.

In addition to the above examples, the most recently reported g-C₃N₄-based S-scheme heterojunctions for H₂O₂ production are displayed in Table 4 [165–174]. It is found that g-C₃N₄ can be used to construct S-scheme heterojunctions with various oxidation and reduction-type photocatalysts and show significantly enhanced activity. For instance, Fang et al. [168] designed a hollow sphere structure Pt/g-C₃N₄/BiOBr S-scheme heterojunction via a solvothermal method. The resultant Pt/g-C₃N₄/ BiOBr heterojunction exhibited a prominent H₂O₂-production rate of 225 μ mol h⁻¹ g⁻¹. Das *et al.* [169] developed a Fe₂O₃ quantum dots/boron-doped g-C₃N₄ (Fe₂O₃ QD/B-g-C₃N₄) Sscheme heterojunction photocatalyst by an *in-situ* generation strategy. The resultant Fe₂O₃ QD/B-g-C₃N₄ heterojunction showed an obviously reinforced H2O2-production rate of 729 μ mol h⁻¹ g⁻¹, nearly two times higher than the pure g-C₃N₄, mainly due to the generation of S-scheme heterojunction between Fe₂O₃ QD and B-g-C₃N₄. Consequently, all the reported g-C₃N₄-based S-scheme heterojunctions displayed excellent photocatalytic H₂O₂ production performance.

Other applications

Except for the above applications of photocatalytic degradation, H_2 evolution, H_2O_2 production, and CO_2 reduction, $g-C_3N_4$ based S-scheme heterojunction photocatalysts have also been used to produce O_2 and fixate nitrogen (N_2). For instance, Li *et al.* [175] constructed a $g-C_3N_4/Ag_3PO_4$ S-scheme heterojunction through an in-situ synthetic strategy, involving the premier fabrication of $g-C_3N_4$ nanotube and the next *in-situ* production of Ag_3PO_4 onto $g-C_3N_4$ (Fig. 19a). The obtained g-



Figure 18 (a) H_2O_2 production rates. (b) Recycling tests. (c) AFM image and the corresponding surface potential maps: (d) dark and (e) light. (f) Surface potentials from A to B and (g) S-scheme mechanism. Reprinted with permission from Ref. [167]. Copyright 2023, Elsevier.

 C_3N_4/Ag_3PO_4 heterojunction composite displayed an excellent photocatalytic O₂-production rate of ca. 370.2 µmol L⁻¹ h⁻¹, over 3-fold superior to the pure Ag₃PO₄ (Fig. 19b). The above enhancements could be mainly due to the fact that the generated g-C₃N₄/Ag₃PO₄ S-scheme heterojunction significantly improved the migration efficiency and redox ability of carriers.

 N_2 fixation technology is essential for the development of industry and agriculture. The proportion of N_2 in the air is as high as 78%. However, most plant corpus cannot directly absorb

and utilize the N_2 in the air. Traditional industrial ammonia synthesis can convert N_2 into ammonia that is easily absorbed by organisms, while it requires high temperature, high pressure, and H_2 energy as a raw material. Therefore, there is an urgent need to develop new, gentle, and green technologies to convert abundant nitrogen into nitrogen that can be absorbed by living organisms. Photocatalytic N_2 -fixation technology is driven by solar energy, N_2 , and water as raw materials; N_2 can be converted into ammonia easily absorbed by organisms. For instance,

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Table 4 Recently reported g-C₃N₄-based S-scheme heterojunction photocatalysts for H₂O₂ evolution

S-scheme heterojunction	Sacrificial agent	Light source (wavelength/nm)	$\begin{array}{c} H_2O_2 \ production \ rate \\ (\mu mol \ h^{-1} \ g^{-1}) \end{array}$	Enhancement factor <i>versus</i> g-C ₃ N ₄	Apparent quantum yield (%)	Ref.
CdS/g-C ₃ N ₄	_	3×50 W Halogen lamp	23,440	2	_	[165]
g-C ₃ N ₄ /Cu ₂ O-Pd	_	500 W Xe lamp ($\lambda > 400$ nm)) 34,000	_	3.3	[166]
Zn-TCPP/g-C ₃ N ₄	Ethanol	300 W Xe lamp	355.1	3.1	7	[167]
Pt/g-C ₃ N ₄ /BiOBr	_	300 W Xe lamp ($\lambda > 400$ nm)) 225	_	_	[168]
Fe ₂ O ₃ QD/B-g-C ₃ N ₄	5% IPA	250 W Hg lamp ($\lambda \ge 420$ nm)) 729	2	_	[169]
S-g-C ₃ N ₄ /TiO ₂	_	300 W Xe lamp (300 $\leq \lambda \leq$ 700 nm)	2128	2.6	0.6	[170]
PCN/MnS	_	300 W Xe lamp	4188	_	8.5	[171]
CN QDs/BiOBr	_	300 W Xe lamp	_	_	_	[172]
NH ₂ -MIL-101(Fe) @MCN/Bi ₂ O ₃	_	300 W Xe lamp ($\lambda > 420$ nm)) 327.8	2.7	_	[173]
g-C ₃ N ₄ /PDA	_	300 W Xe lamp ($\lambda > 350$ nm)) 3801.3	2	2.2	[174]



Figure 19 (a) Synthesis process. (b) O₂ production rates and (c) photocatalytic mechanism. Reprinted with permission from Ref. [175]. Copyright 2022, Elsevier.

Mousavi *et al.* [176] constructed a g-C₃N₄/AgBiS₂ S-scheme heterojunction photocatalyst *via* a solvothermal method. The TEM image (Fig. 20a) shows that AgBiS₂ aggregates were attached to the surface of translucent g-C₃N₄ nanosheets (CNNS). Moreover, the lattice fringes and spacings corresponding to CNNS(002) and AgBiS₂(200) could be clearly observed from its HRTEM image in Fig. 20b. Additionally, the resultant g-C₃N₄/AgBiS₂ heterojunction exhibited prominent N₂-fixation (NH₄⁺ generation) activity of ca. 3780 µmol g⁻¹ L⁻¹, over 2-fold and 3-fold higher than that of pure AgBiS₂ and g-C₃N₄, respectively (Fig. 20c). Moreover, the g-C₃N₄/AgBiS₂ sample emerged excellent stability in the cycle test of nitrogenfixation performance (Fig. 20d). The above excellent N₂-fixation activity and stability were mainly due to the fact that the S-scheme g-C₃N₄/AgBiS₂ heterojunction could efficiently accelerate charge transmission efficiency and enhance its redox ability.

CONCLUSIONS AND OUTLOOK

In summary, single-component photocatalysts cannot simultaneously possess high utilization efficiency of solar energy and strong redox abilities of photoexcited charges, thus usually suffering weak photocatalytic activity [177,178]. The star material $g-C_3N_4$ in the field of photocatalysis is no exception. In this case,



Figure 20 (a) TEM and (b) HRTEM images of CNNS/AgBiS₂. (c) Photocatalytic N_2 fixation activities. (d) Recycling tests and (e) photocatalytic mechanism. Reprinted with permission from Ref. [176]. Copyright 2023, Elsevier.

constructing S-scheme heterojunctions between $g-C_3N_4$ with other semiconductors can simultaneously overcome the typical shortcomings of low light energy utilization, rapid recombination, and weak redox abilities of carriers, thus prominently reinforcing its photocatalytic performance [179]. Hence, this review comprehensively comments on the latest research progress of background, fundamental theory, design and preparation, and characterization strategies of $g-C_3N_4$ -based S-scheme heterojunctions. Additionally, various photocatalytic applications of $g-C_3N_4$ -based S-scheme heterojunctions have been detailly illustrated through example discussion and list comparison, involving photocatalytic H_2 production, CO_2 reduction, H_2O_2 production, pollutant degradation, and others.

Although $g-C_3N_4$ -based S-scheme heterojunctions have made phased progress in controllable construction and enhancement of activity, there are still some bottlenecks to overcome, which are listed as follows.

(1) The dynamic process of charge transfer and the intensity, position, and direction of the built-in electric field are still lacking *in-situ* characterization methods to intuitively reveal. It is well-known that revealing the S-scheme mechanism in depth is the prerequisite for its further development. Therefore, there is

an urgent need to develop new characterization technologies to effectively reveal the mechanism. Meanwhile, it is necessary to strengthen interdisciplinary integration to intuitively and efficiently detect the relevant information of the built-in electric field.

(2) According to the carrier transmission path of S-scheme heterojunction, it is not difficult to conclude that the interfacial electric field is the key factor driving the transmission of photoexcitation charges in S-scheme heterojunctions. However, the current researches for the improvement of $g-C_3N_4$ -based S-scheme heterojunction are primarily concentrating on the traditional cocataltyst modification, element doping, morphology regulation, etc. [180–183], and the influences of the built-in electric field of S-scheme heterojunction on its carrier separation efficiency and photocatalytic activity are rarely studied. Therefore, it is extremely indispensable to investigate the regulation of interfacial built-in electric field of $g-C_3N_4$ -based S-scheme heterojunction and its relationship with photocatalytic activity.

(3) As we all know, there is a certain gap between the current photocatalysis efficiency and the industrial application level, which is the key bottleneck of current photocatalysis technology. Improving the efficiency of photocatalysis technology to a level that can be used industrially is the ultimate goal. Therefore, future research should optimize every condition that can improve the activity of the photocatalytic systems from the inside out. Taking the study of g-C₃N₄-based S-scheme heterojunction as an example, the intrinsic structure (such as morphology, specific surface area, and crystallinity) of g-C₃N₄ and other semiconductors, the heterojunction interface structure, the related interfacial electric field structure, and other factors need to be optimized to break through the bottleneck of performance improvement and reach the level of industrial application.

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g-C₃N₄基S型异质结光催化剂

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摘要随着工业化的蓬勃发展,能源短缺和环境污染日益严重,威胁到 人类的生存.光催化技术因其诸多突出优点和广泛的应用前景被认为 是解决能源和环境危机最有前途的技术之一.在众多光催化剂中,石墨 氮化碳(g-C₃N₄)以其独特的电子结构、较高的热稳定性和突出的光电 活性,在清洁燃料生产和环境净化领域得到广泛应用.然而,单组分 g-C₃N₄与其他光催化剂一样,不可能同时拥有高的太阳能利用效率和 强氧化还原能力的光生电荷,导致其光催化效率较低.幸运的是,g-C₃N₄与另一半导体构建异质结可以同时克服太阳能利用效率低、载流 子重组快、氧化还原能力弱的缺点,从而显著提高其光催化性能.鉴于 目前g-C₃N₄基S型异质结的广泛研究,本文对g-C₃N₄基S型异质结研究 背景、概念提出、基本理论、设计制备、表征方法等方面的最新研究 进展进行了较全面的综述.此外,通过实例讨论和列表比较详细讨论了 g-C₃N₄基S型异质结的各种应用,包括光催化制H₂、还原CO₂、降解污 染物、生产H₂O₂.最后,总结了g-C₃N₄基S型异质结当前的研究进展和 不足,并对未来的研究方向进行了展望.