

Review

2D carbon based nanomaterials for photocatalytic energy conversion and environment remediation reactions

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Abstract

The sustainable development of humanity is facing two major challenges: environmental pollution and energy shortage. Solar light is a sustainable, clean, and inexpensive green energy source. Therefore, the efficient and fast utilization, conversion, and storage of solar energy have attracted increasing attention. Semiconductor photocatalytic technology is centered around the chemical conversion and storage of solar energy. By using photocatalysis to remove toxic and harmful pollutants, it will provide a green and environmentally friendly living space. The conversion of solar energy into hydrogen and other chemical energy sources, such as water splitting and CO₂ conversion, has the potential to solve the energy crisis caused by the increasing depletion of fossil energy. Two-dimensional (2D) carbon based photocatalytic materials have displayed great potential in energy production and environmental remediation due to their unique electronic structure, large specific surface area, and controllable optical properties. This article systematically reviews the latest research progress of 2D carbon based materials such as graphene, carbon nitride, and graphene in photocatalytic hydrogen production, CO₂ reduction, H₂O₂ formation, and pollutant degradation. Strategies for material design and performance optimization are analyzed, and current challenges and future development directions are discussed.

Keywords: Solar energy, photocatalysis, carbon materials, energy conversion, environment remediation

INTRODUCTION

The continuous deterioration of environmental pollution and energy shortages makes it urgent to develop alternative clean energy and green low-carbon environmental remediation technologies^[1-3]. Up to today, solar energy is one of the most widely studied renewable energy among many available energy sources^[4]. Among them, semiconductor photocatalytic technology driven by solar energy can achieve a variety of photochemical reaction processes such as photocatalytic N₂ fixation, photocatalytic reduction of CO₂ to produce low-carbon fuels, and photocatalytic water splitting^[5-8], which exhibits green, clean, sustainable and low cost process in comparison to the traditional energy evolution techniques^[9]. To be specific, photocatalysis is a catalytic process based on light energy driving chemical reactions^[10-13]. Under UV or visible-light irradiation, the holes and electrons generated by the photocatalyst undergo oxidation and reduction reactions with the substances adsorbed on its surface, respectively, thereby achieving the conversion of light energy into chemical energy^[14-17]. As shown in Figure 1, the photocatalytic reaction process generally includes three key steps: (1) The incident photons excite the semiconductor to generate photoinduced electron-hole pairs; (2) The photogenerated charge carriers migrate from the bulk phase to the surface or recombine in the bulk phase; (3) The charge transfer or surface recombination occurs between photogenerated carriers and surface adsorbates. These three key steps are interrelated and affect the total efficiency of photocatalytic reactions. Among them, the recombination of charge carriers is unfavorable for photocatalytic reactions^[18-21].

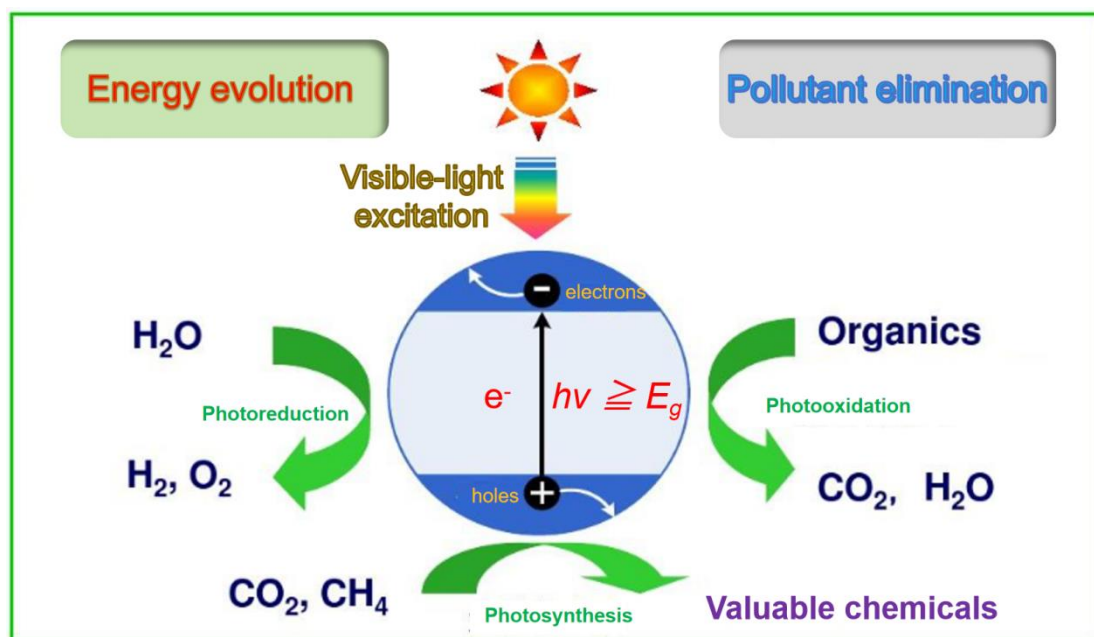


Figure 1. The scheme of photocatalysis technique for energy and environment applications.

Since the discovery of TiO_2 materials for photocatalytic water decomposition in 1972^[22], photocatalytic technology has shown great potential in energy conversion and environmental remediation^[23-25]. However, traditional photocatalyst materials such as TiO_2 , ZnO and SnO_2 have obvious disadvantages including poor visible light response (requiring UV excitation), high carrier recombination rate, and limited specific surface area^[26-28]. The successful preparation of graphene in 2004 opened a new era in 2D material research^[29], and its unique planar structure and excellent electrical properties provided new ideas for photocatalysis^[30]. 2D carbon materials such as graphitic carbon nitride (g- C_3N_4), graphene, MXene, *etc.* have gradually become research hotspots in catalysis and energy fields owing to their fantastic structure and electronic advantages [Figure 2]^[31-35]. Typically, g- C_3N_4 with a bandgap of 2.8 eV, can absorb visible light and has high chemical stability^[36]. Since its application in water splitting in 2009^[37], it has become a representative of metal free photocatalysts. MXene (such as Ti_3C_2), as a novel 2D transition metal carbide, has become an ideal partner for optimizing charge separation due to its high conductivity and specific surface area^[38-40]. In addition, the emergence of new carbon isomers such

as graphitic acetylene has further expanded the diversity of 2D carbon based materials^[41-43]. In a whole, 2D carbon based photocatalysts have become important candidates for solving energy and environmental problems due to their structural advantages and functional tunability.

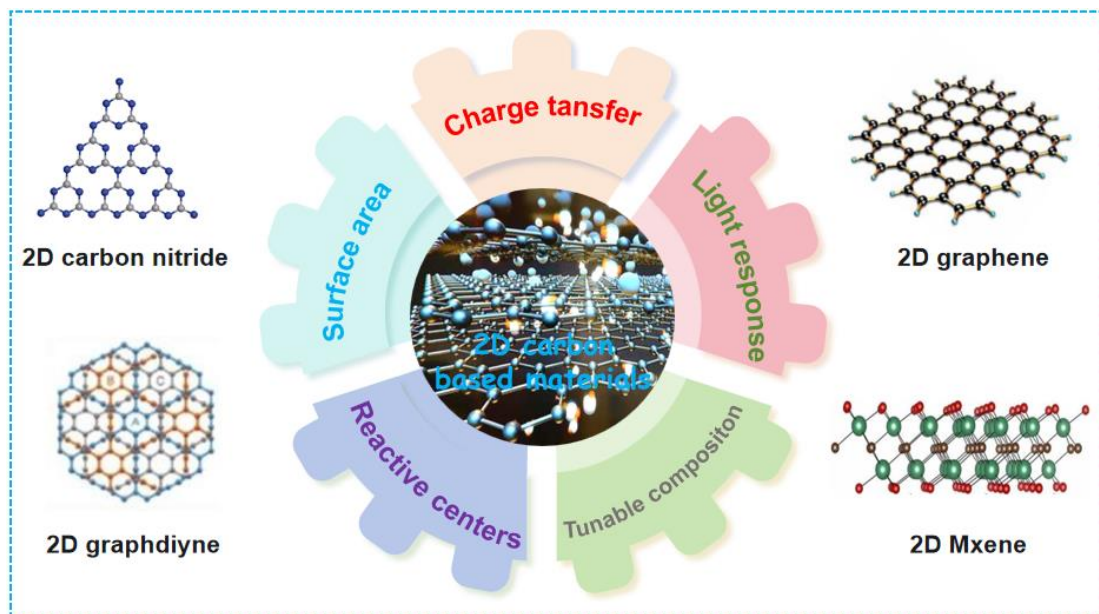


Figure 2. The common two-dimensional carbon based materials and the relevant advantages.

This review article provides a comprehensive overview of the latest research progress in the field of energy generation and environmental remediation using 2D carbon based photocatalysts. It firstly introduces the basic principles of photocatalytic energy generation and environmental remediation, and then illustrates the unique characteristics of carbon based photocatalysts in photocatalytic reactions. Then, the usual modification strategies such as ions doping and defect engineering for enhancing the catalytic activity of 2D carbon based photocatalysts are discussed. Finally, the prospects and obstacle of using 2D carbon based photocatalysts for energy generation and environmental remediation were outlined. We look forward to that this work can present significant advances and knowledge of carbon based photocatalysts design, energy generation, environmental remediation, and activate some new ideas in this promising field.

ADVANTAGES OF 2D CARBON BASED PHOTOCATALYSTS

The advantages of two-dimensional carbon based materials in the field of photocatalysis stem from their unique structural characteristics and synergistic performance effects, which can be summarized from the following aspects including large surface area, rich reactive sites, fast charge separation, tunable light absorption range, low cost of fabrication, *etc.*^[44,45].

Large surface area and rich reactive sites

The high specific surface area and abundant active sites are significant advantages of 2D carbon based materials^[46-48]. The 2D planar structure maximizes the exposure of the material surface, such as biomass derived 2D porous carbon materials with a specific surface area of over 1800 m² g⁻¹, which is much higher than traditional three-dimensional materials. This outstanding characteristics can not only provide sufficient space for substrate adsorption, but also directly serve as catalytic active centers, greatly improving the reaction efficiency. For example, the interlayer gaps of carbon nitride can efficiently adsorb organic pollutants, moreover, its surface functional groups (including - OH, - COOH) and defect sites can act as active reaction centers^[48].

Facile charge separation and shift

The layered structure of two-dimensional materials can shorten the transmission path of photo generated charge carriers and reduce the probability of recombination during migration^[49]. Taking g-C₃N₄ as an example, its planar π - electron conjugated system can achieve rapid delocalization of electrons. When constructing 2D/2D heterojunctions with conductive 2D materials such as graphene and MXene, the efficient charge transfer channels formed at the interface can further accelerate the separation of electron hole pairs. The charge separation efficiency of some composite materials is more than three times higher than that of single materials, providing sufficient active carriers for photocatalytic reactions^[50].

Tunable light absorption range

Tunable light absorption performance can be achieved through structural design of 2D carbon materials^[51]. By doping methods, defect creation, and heterostructure construction, the bandgap width of carbon based photocatalysts can be further narrowed or defect energy levels can be introduced to broaden the visible light responsive range. For example, after constructing heterostructure on semiconductor materials such as titania, the visible light absorption range is significantly improved, exhibiting stronger activity in dye degradation and hydrogen evolution reactions^[52].

Ideal chemical stability

Carbon based materials often have excellent corrosion resistance and thermal stability, and can maintain structural integrity in complex reaction environments such as strong acids, strong bases, and high temperatures^[53]. Typically, sulfur doped g-C₃N₄ shows no significant degradation of catalytic performance after 20 hours in the continuous reaction of phenol degradation^[54]. This stability eliminates the need for frequent replacement of photocatalyst and reduces maintenance costs in practical applications.

Low cost and facile synthesis

The preparation of some two-dimensional carbon based materials has significant cost advantages, especially those using biomass (such as straw, walnut shells, sawdust) as raw materials, which are widely sourced and renewable, and the preparation process conforms to the concept of green chemistry^[55]. For example, g-C₃N₄ photocatalyst can be obtained by simple calcination of inexpensive precursors such as urea and melamine, without the need for complex equipment, and has great potential for large-scale preparation, laying the foundation for its industrial application^[56].

MODIFICATION STRATEGIES OF 2D CARBON BASED PHOTOCATALYSTS

Although photocatalysis has huge application potentials in the fields of energy and

environment, the vast majority of photocatalysts are limited by different factors such as limited visible light absorption range and rapid photogenerated carrier recombination rate, resulting in low photocatalytic activity and efficiency^[57]. Therefore, scientists have been committed to solving two core problems in the photocatalysis reactions,: (1) expanding the visible-light absorption range of photocatalysts; (2) improve the separation efficiency of photoinduced charge carriers. Up to now, the modification methods mainly include: heterostructure construction, ion doping, defect engineering, noble metal deposition, and pore engineering [Figure 3]^[58].

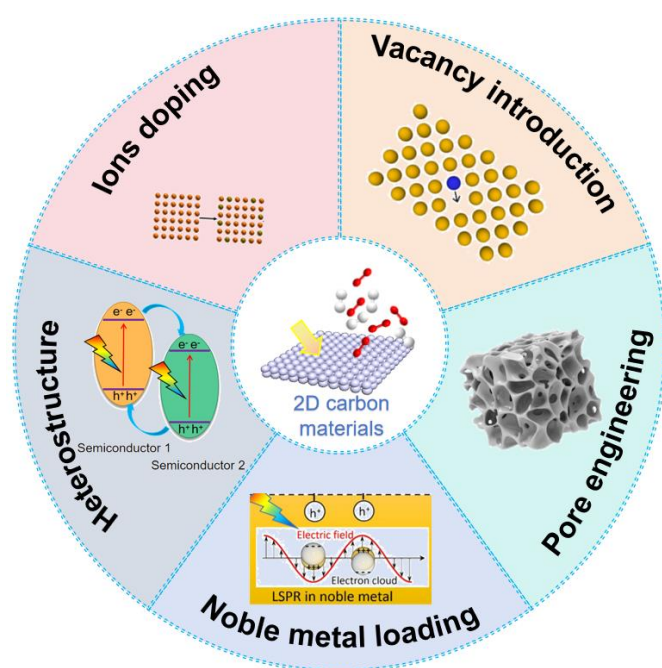


Figure 3. The usual modification strategies of 2D carbon based materials for efficient photocatalytic activity.

Heterojunction construction

The type II, Z-type, and P-N type heterojunctions have been often reported to optimize carrier separation efficiency through interfacial electronic structure regulation^[59-61], but their charge transfer pathways and band alignment mechanisms are different. The type II heterojunction relies on the staggered band alignment of two semiconductors, where photogenerated electrons transfer from the CB of the

semiconductor A to the CB of the semiconductor B, while holes migrate in the opposite direction, achieving spatial carrier separation. However, the redox capability of electrons and holes is weakened [Figure 4a]. The Z-type heterojunction enables the recombination of CB electrons from the semiconductor B with valence band holes from semiconductor A [Figure 4b]. This retains the strong redox-active valence band holes and conduction band electrons of both materials, balancing separation efficiency with reaction activity, and is well-suited for advanced redox reactions (e.g., CO₂ reduction, water splitting). The P-N type heterojunction is based on the contact potential difference between p-type and n-type semiconductors, forming a depletion layer and built-in electric field that drives electrons from the n-type to the p-type while holes migrate in the opposite direction [Figure 4c]. This results in strong carrier separation dynamics and low interfacial charge transfer resistance. Overall, all three types address the critical bottleneck of rapid carrier recombination in photocatalysis through interfacial engineering, while differences in band structure design, charge transfer direction, and redox capability determine their suitability for various photocatalytic applications.

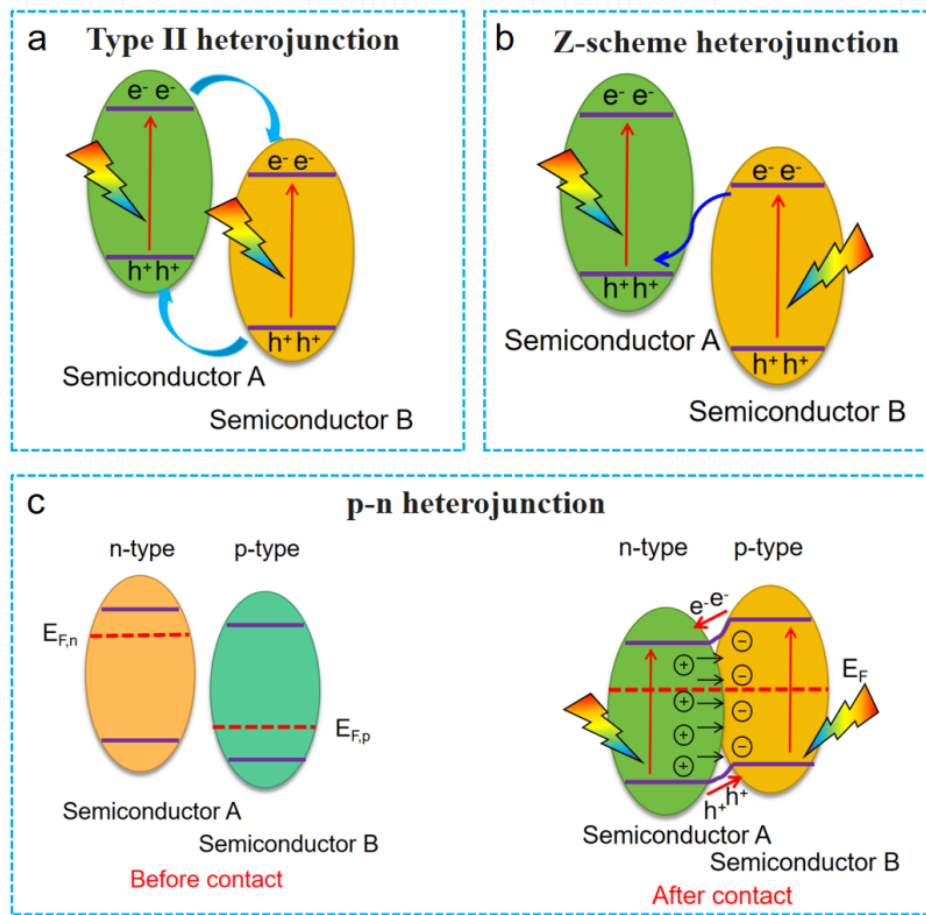


Figure 4. The usual types of heterojunctions, including (a) Type II; (b) Z-type; and (c) P-N type for efficient photocatalytic capability.

Ions doping

Ions doping is one of the most commonly used modification strategies^[62], which involves introducing heterogeneous elements (metal/non-metal/rare-earth elements) into the lattice or surface of semiconductor photocatalysts to alter their electronic structure and band positions, thereby optimizing their light absorption and charge separation performance. For example, when non-metal ions (such as S^{2-} , P^{5+} , B^{3+} , etc.) are introduced into the lattice of C_3N_4 photocatalyst, new impurity energy levels (located between the conduction and valence bands) are introduced^[63], reducing the energy required for photoexcitation. Meanwhile, metal ions can act as electron traps to suppress the recombination of photogenerated electron hole pairs^[64].

Defect engineering

Defect engineering is the process of introducing intrinsic defects (such as oxygen vacancies, metal vacancies, lattice distortions) into photocatalysts through physical methods (such as plasma treatment, ball milling) or chemical methods (such as hydrogen reduction, vacancy compensation)^[65]. Defects can serve as active sites or charge transport channels while adjusting the band structure. Typically, unsaturated atomic coordination at the defect site enhances the adsorption capacity for substrates such as pollutants, H₂O, and CO₂ (e.g. oxygen vacancies can adsorb CO₂ molecules through lone pair electrons), thus accelerating photocatalytic reaction^[66].

Pore engineering

After introducing the porous structure in photocatalysts, the catalytic performance can be obviously improved due to efficient mass transfer efficiency and rich reactive sites. The biggest advantage of porous structures, especially micropores and mesopores, is their extremely high specific surface area^[67]. For example, the specific surface area of porous carbon and MOFs (metal organic frameworks) based photocatalysts can reach hundreds or even thousands of m² g⁻¹, far higher than those without pores or dense structures. Increasing the specific surface area can directly expose more active sites, improve the probability of contact between reactants and active sites, and thus enhance photocatalytic efficiency. Moreover, reactants need to diffuse to the surface of the catalyst in photocatalytic reactions. The products must be detached from the surface in a timely manner to avoid occupying active sites. The porous structure of the photocatalysts can effectively solve the problem of mass transfer obstruction^[68].

Noble metal loading

Noble metal nanoparticles (such as Au, Ag, *etc.*) with size of 1-50nm can be modified onto the surface of photocatalysts through physical adsorption or chemical bonding, which mainly utilize surface plasmon resonance (SPR) effect and charge trap effect to enhance activity of photocatalysts^[69]. Under visible light irradiation, surface free electrons of noble metal nanoparticles undergo collective oscillation upon excitation,

resulting in strong visible light absorption (such as Au nanoparticles having an SPR absorption peak around 520 nm), which can broaden the light response range to visible light or even near-infrared light^[70]. Moreover, the Fermi level of noble metals is lower than the bottom of the conduction band of most semiconductor photocatalysts, and the photogenerated electrons generated by photocatalysts would transfer to the surface of noble, suppressing electron-hole recombination.

APPLICATIONS OF 2D CARBON MATERIALS

2D carbon based photocatalysts have unique advantages due to their atomically thin structure, large specific surface area, abundant active sites, and promoted adsorption of reactants^[71]. The interlayer short-range path can suppress the recombination of photogenerated charge carriers and improve separation efficiency. At the same time, the adjustable electronic structure is adaptable to multiple reactions with a wide light response range (from ultraviolet to visible light). Through ions doping, heterostructure building and other modifications, the performance can be continuously optimized, and there is enormous potential in the fields of energy conversion and environmental remediation^[72]. In this chapter, the representative 2D carbon based photocatalysts involving g-C₃N₄, graphene, and MXene for photocatalytic water splitting, CO₂ conversion, H₂O₂ generation, and pollutant elimination were discussed in depth.

Photocatalytic water splitting

Photocatalytic water splitting is a green energy evolution technology that utilizes semiconductor materials to decompose water into hydrogen and oxygen under light illumination^[73]. When photocatalysts absorbing photons with sufficient energy, the electron-hole pairs can be generated. The electrons reduce water to generate H₂, while holes oxidize water to release O₂. This process simulates natural photosynthesis, which can convert solar energy into chemical energy and produce clean hydrogen product^[74]. However, some challenges including low catalytic efficiency and insufficient catalyst stability are presented, current research focuses on developing narrow bandgap materials especially 2D carbon based materials to optimize water

splitting performance, which is expected to achieve carbon neutrality goals.

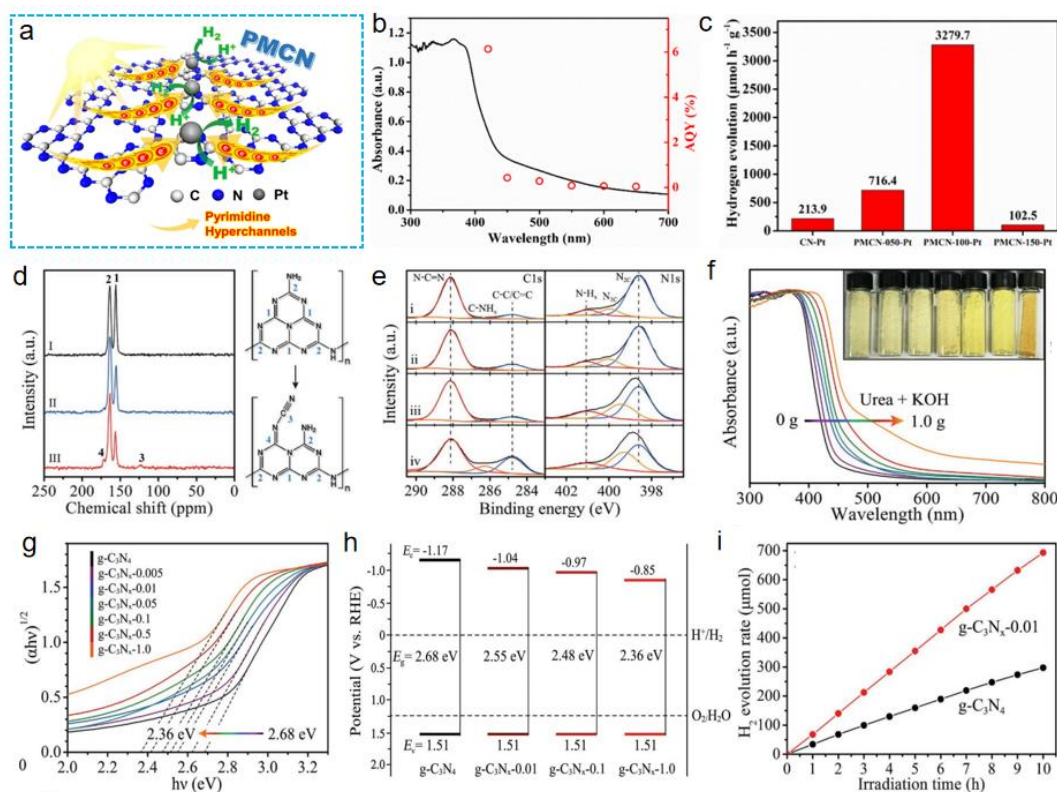


Figure 5. (a) The scheme of photoexcited electron transfer channel of Pt modified 2D g-C₃N₄ for efficient photocatalytic hydrogen evolution; (b) UV-visible absorption spectrum and (c) hydrogen evolution rates of the obtained photocatalysts. Reproduced with permission.^[77] Copyright 2018, Elsevier. The (d) NMR; (e) XPS; (f) UV-visible absorption spectra; and (g) band gap width value of the obtained C₃N₄ photocatalysts. (h) The precise electronic bandgap configuration, and (i) hydrogen evolution rates of the obtained C₃N₄ photocatalysts. Reproduced with permission.^[78] Copyright 2017, Wiley.

Wang's group firstly reported that g-C₃N₄ can be used as a stable photocatalyst to split water to produce hydrogen in 2009. Since then, g-C₃N₄ has received increasing attention by researchers^[75]. As a carbon based polymer semiconductor, different modification strategies have been developed on g-C₃N₄ for improved water splitting performance^[76]. For example, Shi's team constructed a photoexcited electron transfer

channel through loading platinum (Pt) on pyrimidine modified 2D g-C₃N₄^[77], which can significantly improve the photocatalytic water splitting performance. To be specific, the introduced pyrimidine groups act as electron acceptors into the nitrogen doped g-C₃N₄ skeleton, while Pt nanoparticles function as electron donors, thus endowing an efficient electron transfer pathway [Figure 5a]. This structure optimizes the separation and migration efficiency of photogenerated charge carriers, expanding the visible light absorption range to 600 nm [Figure 5b] and achieving a hydrogen production rate 12 times that of the pristine g-C₃N₄ photocatalyst [Figure 5c]. The experimental characterization and theoretical calculations confirmed that the synergistic effect of pyrimidine Pt reduces the reaction energy barrier, providing great prospect for water splitting.

Zhang's group proposed an innovative strategy for synthesizing nitrogen deficient g-C₃N₄ sample through alkaline assisted pyrolysis (such as NaOH, KOH)^[78]. It was revealed that the introduction of alkali not only promotes the volatilization of precursors (such as urea or melamine) during pyrolysis, thereby creatively introducing nitrogen vacancy defects in the g-C₃N₄ [Figure 5d and e], but also effectively regulates the band structure of the material. These nitrogen defects act as active sites, significantly enhancing the absorption of visible light [Figure 5f-h] and greatly suppressing the recombination of photo generated electron hole pairs. Finally, this optimized nitrogen deficient g-C₃N₄ catalyst exhibited significantly higher photocatalytic activity for water splitting and hydrogen production under visible light irradiation than pristine g-C₃N₄ catalysts [Figure 5i].

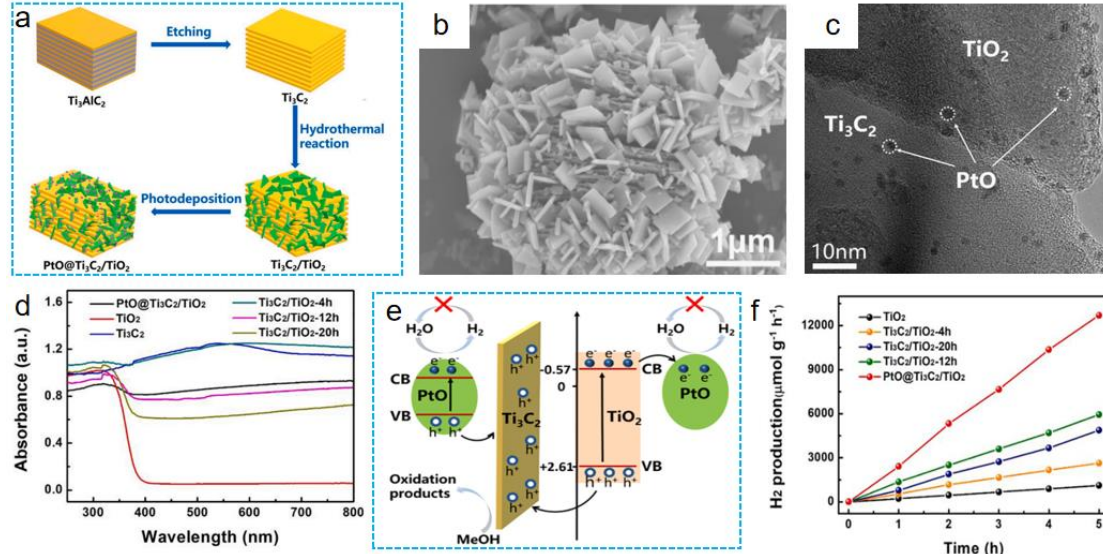


Figure 6. (a) The schematic synthesis process of $\text{Ti}_3\text{C}_2/\text{TiO}_2$ composite photocatalyst. (b) SEM and (c) TEM images the obtained photocatalysts; (d) The UV-visible absorption spectra of the obtained $\text{Ti}_3\text{C}_2/\text{TiO}_2$ composite photocatalysts; (e) The electronic bandgap configuration of the $\text{Ti}_3\text{C}_2/\text{TiO}_2$ heterostructure composite for hydrogen production; and (f) hydrogen evolution rates of the obtained $\text{Ti}_3\text{C}_2/\text{TiO}_2$ composite photocatalysts. Reproduced with permission.^[81] Copyright 2021, Elsevier.

MXenes materials have several excellent advantages, including atomically thin 2D morphology, metallic conductivity, hydrophilic surface, and high specific surface area, which was often used for photocatalytic water splitting^[79,80]. Typically, Su's team developed a $\text{Ti}_3\text{C}_2/\text{TiO}_2$ composite photocatalyst by in-situ conversion of Ti_3C_2 MXene modified with PtO nanodots for efficient photocatalytic hydrogen production [Figure 6a]^[81]. In this composite, Ti_3C_2 is partially converted into TiO_2 nanosheets, while ultrafine PtO nanodots are anchored on the surface as co-catalysts. This unique structure not only retains the high conductivity of MXene, but also forms a tight $\text{Ti}_3\text{C}_2/\text{TiO}_2$ heterojunction, significantly promoting the separation and transport of photogenerated charges [Figure 6b and c]. In addition, the composite photocatalyst exhibits enhanced photoresponse in the visible light region [Figure 6d]. Mechanism studies demonstrated that PtO nanodots act as electron traps and reactive active sites, synergistically enhancing the photocatalytic performance with the conductive network

of Ti_3C_2 [Figure 6e]. Photocatalysis experiments illustrated that the hydrogen production rate of the 2D $\text{Ti}_3\text{C}_2/\text{TiO}_2$ composite is 15 times that of pure TiO_2 [Figure 6f].

In general, 2D carbon based photocatalysts have shown great potential in the field of photocatalytic water splitting for hydrogen production. Through strategies such as heterojunction construction, defect engineering, and element doping, the efficiency of photogenerated carrier separation and visible infrared light response range have been significantly improved. Future research needs to focus on low-cost and large-scale preparation adaptability to complex environments, and device integration to promote the practical application of solar driven hydrogen production.

Photocatalytic H_2O_2 evolution

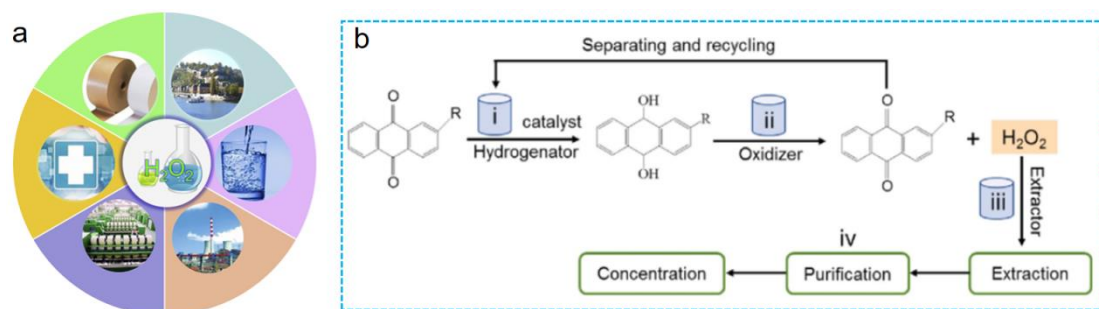


Figure 7. (a) The applications of H_2O_2 in different fields; (b) The scheme of traditional anthraquinone technique for H_2O_2 synthesis. Reproduced with permission.^[82] Copyright 2022, Elsevier.

H_2O_2 as a high-value chemical, has a wide range of applications in clean fuels, wastewater treatment, disinfection and cleaning, organic synthesis, and biological therapy [Figure 7a]^[82]. Photocatalytic hydrogen peroxide (H_2O_2) synthesis is considered a green and sustainable method that utilizes solar energy to drive the reaction of oxygen and water on the photocatalyst surface to generate H_2O_2 ^[83]. This process usually uses semiconductor materials (such as C_3N_4 , TiO_2 , *etc.*) as

photocatalysts, and O_2 is reduced in two steps to H_2O_2 ($O_2 + 2H^+ + 2e^- \rightarrow H_2O_2$) through photogenerated electrons, while holes oxidize water or sacrificial agents to maintain total charge balance. Compared to traditional anthraquinone technique [Figure 7b], photocatalytic H_2O_2 synthesis has milder conditions, lower energy consumption, and negligible pollution^[82,84]. 2D carbon based materials with excellent structure, morphology and electronic properties have been widely utilized as photocatalyst to synthesize H_2O_2 in environmental conditions.

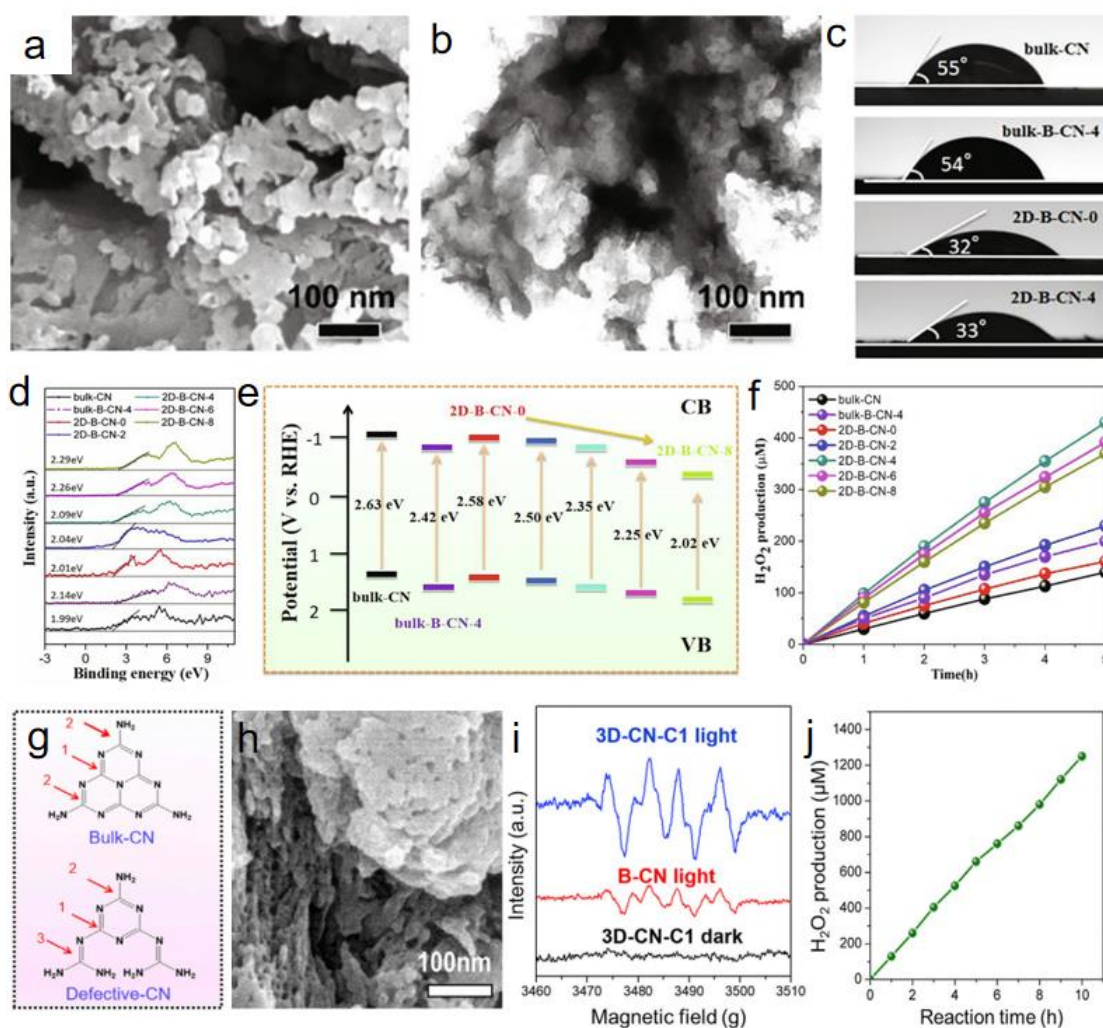


Figure 8. The (a) SEM, (b) TEM, and (c) water contact angle images of the prepared boron doped g- C_3N_4 nanoplates; (d) XPS valence band spectra and (e) specific electronic band gap structures of the obtained boron doped g- C_3N_4 nanoplates. (f) The photocatalytic hydrogen evolution rates of the obtained 2D C_3N_4 photocatalysts. Reproduced with permission.^[85] Copyright 2022, Elsevier; (g) The basic structure of

the bulk and defective C_3N_4 samples; The (h) SEM, (i) ESR spectra and (j) time dependent photocatalytic H_2O_2 evolution of the prepared 2D deficient C_3N_4 photocatalyst. Reproduced with permission.^[86] Copyright 2022, Elsevier.

Ding and co-workers developed a hydrophilic boron doped g- C_3N_4 nanoplates [Figure 8a and b] to achieve efficient photocatalytic H_2O_2 synthesis^[85]. Experimental results suggested that boron doping not only improves the hydrophilicity of the 2D g- C_3N_4 nanoplates [Figure 8c] but also regulates the electronic structure to simultaneously promote oxygen reduction reaction [Figure 8d and e]. Through boron atom doping and morphology control, the g- C_3N_4 nanoplates displayed enhanced visible light absorption, optimized charge separation efficiency, and abundant surface active sites. The above properties facilitate the H_2O_2 synthesis in water phase. As expected, the boron doped g- C_3N_4 nanoplates achieved 5 times higher H_2O_2 yield than that of bulk g- C_3N_4 sample through a two electron oxygen reduction pathway [Figure 8f]. In another work, their team developed a new strategy for efficient photocatalytic synthesis of H_2O_2 by constructing carbon vacancies in porous g- C_3N_4 nanoplates [Figure 8g and h]^[86]. By introducing carbon vacancy defects into the g- C_3N_4 framework, the electronic structure of the photocatalyst is effectively regulated, promoting the generation and stability of superoxide radicals ($\bullet O_2^-$), which are deemed as essential species during H_2O_2 formation [Figure 8i]. It was revealed that the formed carbon vacancy defects significantly improve the selectivity of photocatalytic two electron oxygen reduction reaction, resulting in a 8.3 times greater H_2O_2 formation rate than that of the pristine g- C_3N_4 [Figure 8j]. Theoretical calculations confirm that carbon vacancies as active sites optimize the O_2 adsorption activation process while suppressing the decomposition of H_2O_2 . The above cases of H_2O_2 evolution via 2D g- C_3N_4 sample provides a promising solution for solar energy conversion.

Mishra's group designed a ternary MXene heterojunction photocatalyst (MXene/ Bi_2WO_6 /g- C_3N_4) for synergistically enhancing the synthesis of H_2O_2 ^[87]. An

efficient charge transfer channel was constructed by combining conductive MXene with $\text{Bi}_2\text{WO}_6/\text{g-C}_3\text{N}_4$. In this ternary system, MXene can act as an electronic medium, which not only promotes carrier separation through Schottky junction, but also optimizes the interfacial reaction kinetics. Experimental and theoretical calculations indicated that the ternary MXene/ $\text{Bi}_2\text{WO}_6/\text{g-C}_3\text{N}_4$ system retains strong redox ability through the Z-type mechanism, and the introduction of MXene further suppresses charge recombination. As a result, the ternary MXene/ $\text{Bi}_2\text{WO}_6/\text{g-C}_3\text{N}_4$ photocatalyst give rise to an efficient H_2O_2 yield of 1.78 mM h^{-1} , being about 3.2 times greater than that of binary $\text{Bi}_2\text{WO}_6/\text{g-C}_3\text{N}_4$ system. Hou et al. developed a 2D graphene oxide (GO)-based photocatalyst for efficient synthesis of H_2O_2 driven by solar light radiation in the absence of organic electron donors^[88]. It was confirmed that the unique electronic structure and surface oxygen-containing functional groups of GO enable it to directly utilize water and oxygen, synergistically producing H_2O_2 through dual channels (two electron water oxidation and two electron oxygen reduction) without the need for any sacrificial agents. Mechanism studies indicated that the carboxyl and epoxy groups of GO serve as active sites, while promoting the separation of photogenerated charges and surface reaction kinetics. Under simulated sunlight irradiation, the generation rate of H_2O_2 reaches $176 \mu \text{ M h}^{-1}$, far exceeding most traditional semiconductor photocatalysts.

2D carbon based photocatalysts have shown outstanding advantages in photocatalytic synthesis of H_2O_2 due to their high specific surface area, tunable electronic structure, and wide photoresponse range. In recent years, strategies such as heterostructure construction and atomic doping have significantly improved carrier separation efficiency and oxygen reduction selectivity. To promote practical application of this technique, it is necessary to focus on precise structural control to enhance light absorption and charge separation, optimize reaction systems to suppress side reactions, and develop low-cost and large-scale preparation processes.

Photocatalytic CO_2 conversion

The excessive emission of carbon dioxide (CO_2) has led to global issues including higher temperatures, rising sea levels, and loss of species diversity, posing severe challenges to sustainable development^[89]. Therefore, the capture, storage, and conversion of CO_2 have received widespread attention from scientists. Compared with traditional, high-energy capture or geological storage methods, realizing the resource utilization of CO_2 is a more promising approach that can simultaneously alleviate greenhouse effect and energy crisis^[90,91]. Photocatalytic

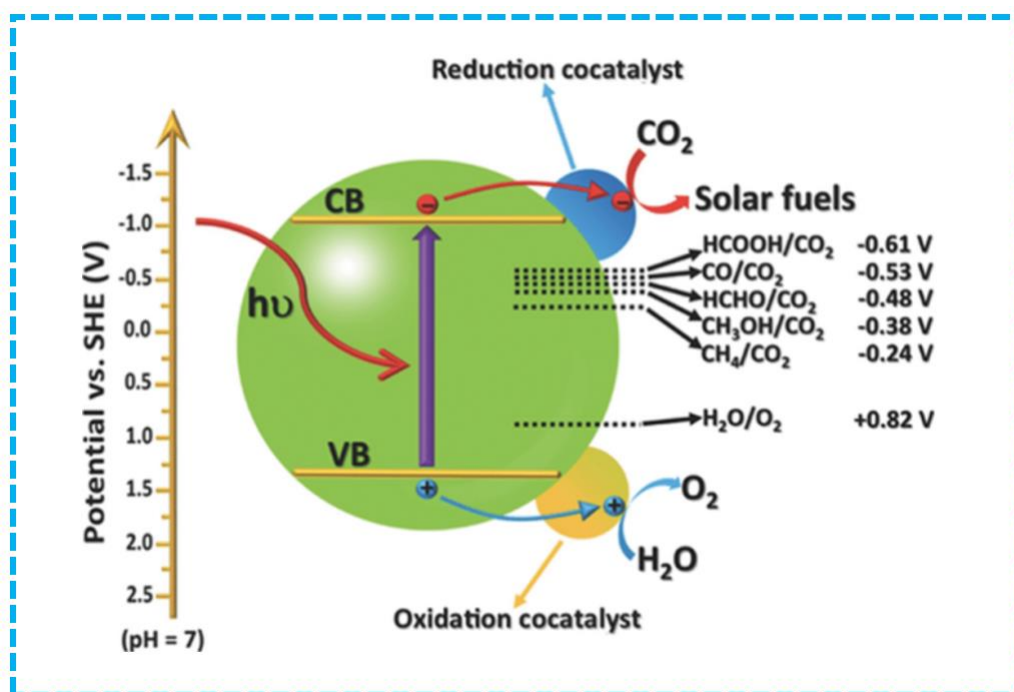


Figure 9. The scheme of photocatalytic conversion of CO_2 to produce different hydrocarbon fuels. Reproduced with permission.^[92] Copyright 2021, Wiley.

conversion of CO_2 to produce hydrocarbon fuels is considered one of the most promising strategies for achieving carbon neutrality goals. The high value-added fuels or chemicals such as CO , CH_4 and CH_3OH can be produced by photocatalytic CO_2 conversion [Figure 9], which is considered as a green and low-cost technology that utilizes solar energy to drive the reduction of CO_2 ^[92]. Under UV or visible light excitation, the photogenerated electrons of photocatalysts can reduce CO_2 molecules

to generate different products, while holes oxidize water or sacrificial agents to provide protons. The great challenges of photocatalytic CO₂ conversion lie in the chemical inertness of CO₂ molecules, selective generation of target products, and low efficiency of light-chemical energy conversion^[93,94]. 2D carbon based materials such as g-C₃N₄ and MXene attracted growing attention in CO₂ conversion field due to their high surface area and excellent charge separation and transfer capabilities. Moreover, the thinness of these materials shortens the carrier transport path, improves the adsorption and activation of CO₂ and water, reduces energy barriers, and selectively enhances specific reactions.

Typically, Wang's team reported an efficient photocatalytic CO₂ reduction system using boron doped g-C₃N₄ (BCN) modified CdS nanoparticles [95]. The decoration of CdS nanoparticles onto BCN can generate a heterojunction with good visible light response ability [Figure 10a]. Moreover, a variety of characterizations suggested that the heterojunction structure significantly enhances the separation efficiency of photogenerated charge carriers [Figure 10b and c] and optimizes the adsorption and activation ability of CO₂ molecules, which can efficiently converts CO₂ into carbon hydrogen fuels such as CO and CH₄ under visible light irradiation. The activity of BCN@CdS composite is significantly improved compared to the single CdS sample [Figure 10d-f]. Similarly, Pei *et al.* achieved controllable CO₂ to syngas (CO/H₂) conversion by constructing an S-type heterojunction ZnGa₂O₄/g-C₃N₄ photocatalyst [Figure 10g]^[96]. Through strong electronic coupling, this ZnGa₂O₄/g-C₃N₄ composite system not only significantly

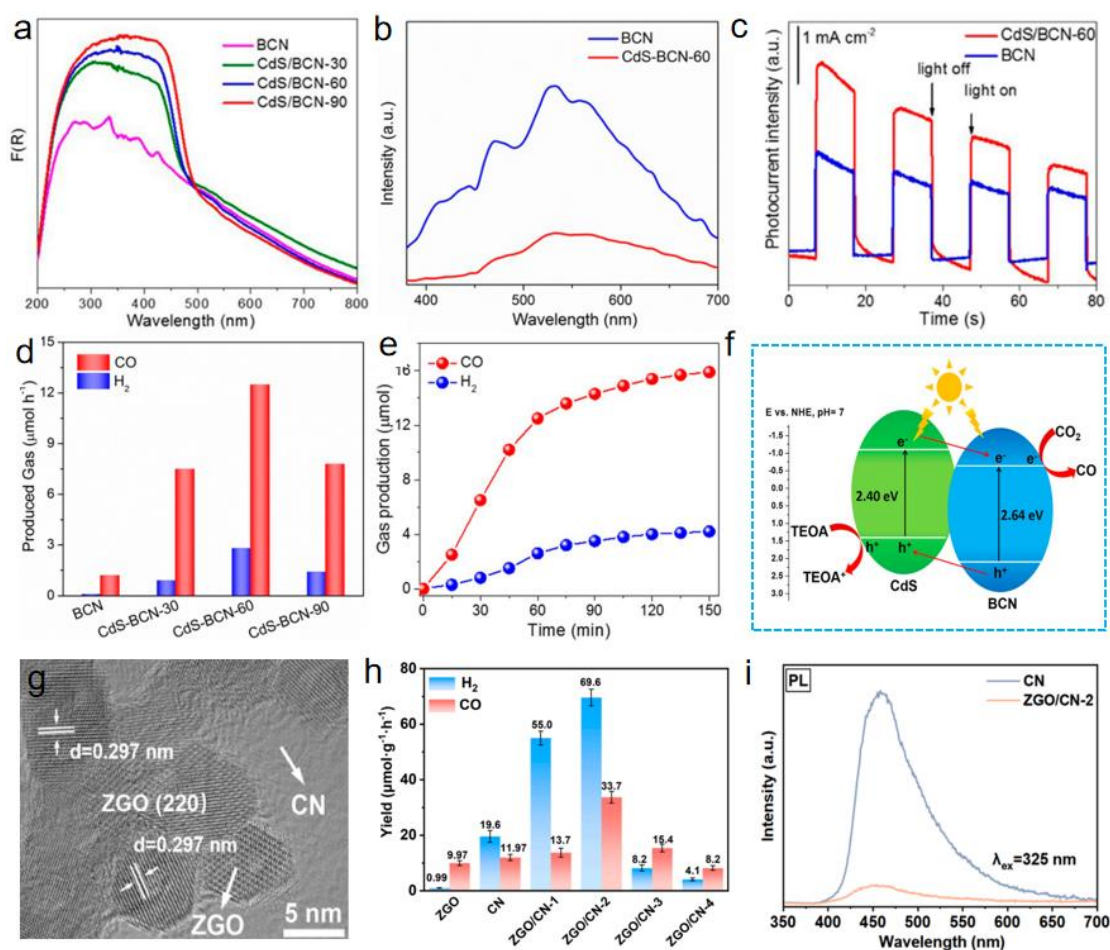


Figure 10. The (a) UV-Visible absorption; (b) PL spectra; and (c) photocurrent response curve of the prepared BCN@CdS composite photocatalysts. The produced CO and CH₄ via photoreduction of CO₂ over different BCN@CdS composite; (d) Time dependent CO and CH₄ evolution over the photocatalyst; (f) The schematic heterostructure configuration of the obtained BCN@CdS composite; (g) The TEM image of the ZnGa₂O₄/g-C₃N₄ samples; (h) The produced CO and CH₄ via photoreduction of CO₂ over different ZnGa₂O₄/g-C₃N₄ samples; and (i) PL spectra of the prepared C₃N₄ photocatalyst. Reproduced with permission.^[95] Copyright 2018, American Chemical Society.

promotes the spatial separation of photogenerated carriers, but also achieves precise control of the CO/H₂ ratio in the product within the range of 0.9-4.3 [Figure 10h]. It was confirmed that the S-scheme charge transfer pathway formed between ZnGa₂O₄ and g-C₃N₄ effectively preserves highly reducing electrons and oxidizing holes

[Figure 10i], while the interface Ga-N bond serves as an electron bridge to enhance the interaction. Under visible light excitation, the $\text{ZnGa}_2\text{O}_4/\text{g-C}_3\text{N}_4$ photocatalyst exhibits excellent CO_2 reduction activity (CO yield of $28.7 \mu\text{mol g}^{-1} \text{h}^{-1}$) and ideal stability.

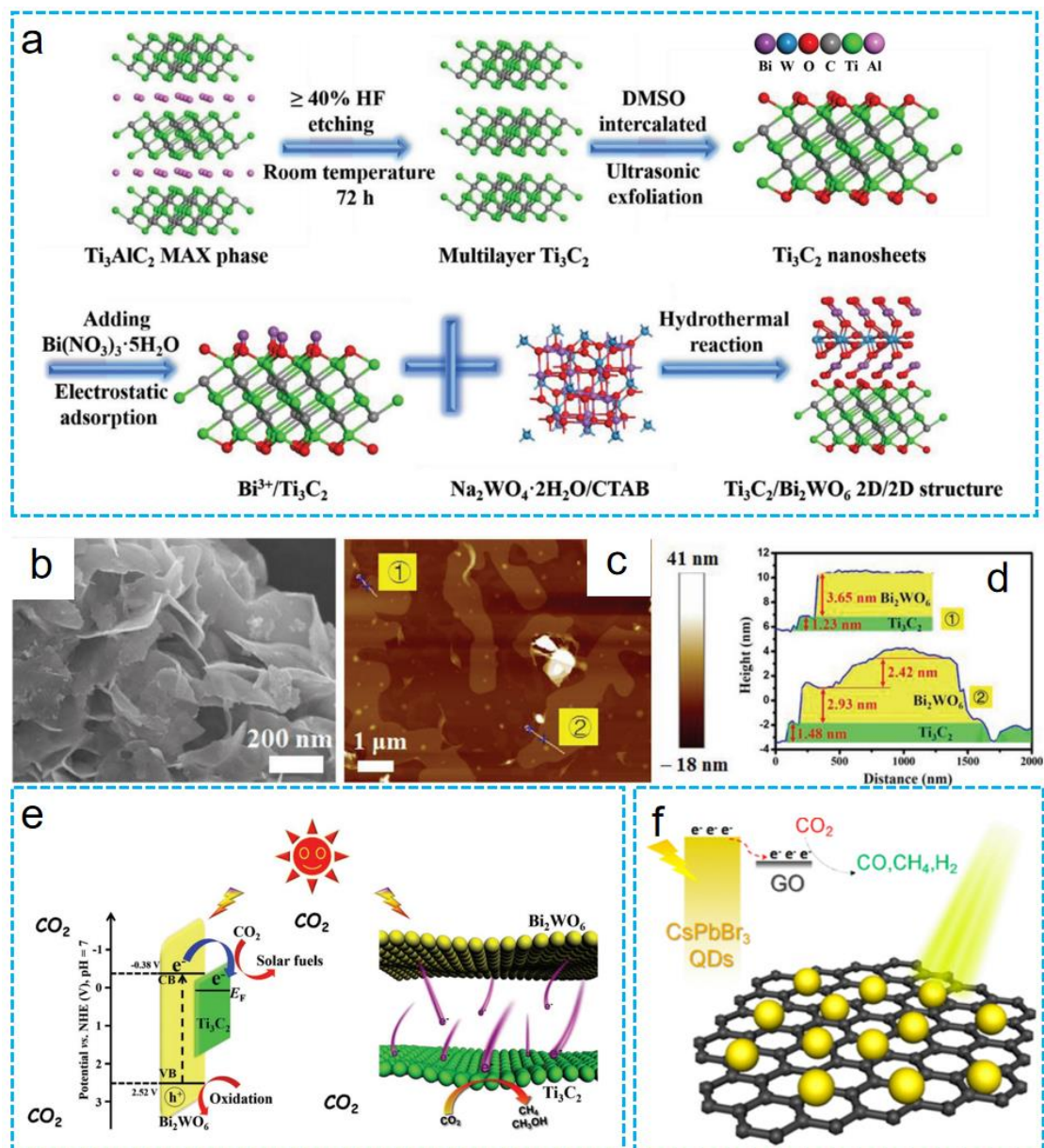


Figure 11. (a) The schematic preparation process of the 2D ultrathin MXene/ Bi_2WO_6 photocatalysts; (b) SEM; (c) AFM; and (d) thickness profile image of the 2D ultrathin MXene/ Bi_2WO_6 photocatalysts; (e) Photocatalytic CO_2 reduction mechanism over the 2D heterostructure photocatalyst. Reproduced with permission.^[99] Copyright 2018, Wiley; (f) Photocatalytic CO_2 reduction scheme over the CsPbBr_3 PQDs loaded GO

photocatalyst. Reproduced with permission.^[102] Copyright 2017, American Chemical Society.

MXene has the extraordinary characteristics such as atomically thin 2D morphology, metallic conductivity, and high specific surface area, which can effectively adsorb and activate CO₂ molecules, providing abundant active sites for photocatalytic reduction of CO₂^[97-98]. For example, Yu et al. constructed a 2D/2D heterojunction of ultra-thin MXene (Ti₃C₂T_x)/Bi₂WO₆ nanosheets [Figure 11a] for efficient photocatalytic CO₂ reduction^[99]. A tight interface contact was formed by combining MXene with Bi₂WO₆ nanosheets with atomic layer thickness [Figure 11b-d], which can significantly promote the separation and migration of photogenerated charges. Mechanism studies confirmed that 2D ultra-thin MXene not only acts as an electron acceptor to promote charge transfer, but also enhance CO₂ adsorption and activation due to its abundant surface functional groups [Figure 11e]. As a result, photocatalytic experiments displayed that the heterojunction catalyst exhibits excellent CO₂ reduction performance under visible light, with a CO yield of up to 28.6 μmol g⁻¹ h⁻¹, which is 4.3 times that of pure Bi₂WO₆ photocatalyst. In another study, the researchers developed a surface alkalized Ti₃C₂ MXene as an efficient co-catalyst^[100], significantly improving the photocatalytic CO₂ reduction performance of ZnO. The introduction of -OH/-O⁻ functional groups on the surface of Ti₃C₂ MXene through NaOH treatment not only improved its interface contact with ZnO, but also enhanced its CO₂ adsorption and activation ability. The Ti₃C₂/ZnO composite catalyst achieved a CO yield of 12.5 μmol g⁻¹ h⁻¹ under visible light excitation, which is 8 times higher than that of pure ZnO photocatalyst. Mechanism analysis demonstrated that MXene with surface alkalization acts as an electron acceptor to promote charge separation, while its alkaline sites lower the CO₂ reduction energy barrier, thus leading to higher CO₂ reduction efficiency.

Just like 2D MXene and g-C₃N₄ samples, 2D graphene composite as efficient photocatalyst was often used for CO₂ reduction reaction owing to their fantastic

structure and electronic advantages^[101]. For example, the researchers reported a CsPbBr₃ perovskite quantum dot (CsPbBr₃ PQDs)/graphene oxide (GO) composite material for efficient photocatalytic CO₂ reduction^[102]. The efficient visible light response was achieved by anchoring CsPbBr₃ PQDs on the surface of GO. Photocatalytic results suggested that the CsPbBr₃ PQDs/GO photocatalyst efficiently converts CO₂ into CO and CH₄ under visible light excitation, and its catalytic activity is 3.5 times that of pure CsPbBr₃ PQDs. Mechanism analysis demonstrated that GO not only serves as an electronic conduction network to promote charge separation, but also enhance the adsorption capacity of CO₂ owing to its oxygen-containing functional groups [Figure 11f]. In a whole, the excellent light capture properties of CsPbBr₃ PQDs component with the excellent charge transfer ability of GO synergistically enhance the photocatalytic CO₂ reduction performance.

In a whole, 2D carbon based photocatalysts have broad prospects in the field of photocatalytic CO₂ reduction. By different modification methods, the CO₂ reduction efficiency significantly improves by virtue of the photogenerated charge separation efficiency, CO₂ adsorption activation ability, and product selectivity. In the future study, the multi-level in-situ characterization techniques to analyze reaction mechanisms and machine learning assisted material design should be concerned to promote the practical application of solar driven CO₂ resource utilization.

Organics degradation

With the rapid development of industrialization, organics pollution has become increasingly serious, posing a serious threat to environment and human health^[103]. Traditional organic pollutant treatment technologies including adsorption, precipitation, and biodegradation pose a variety of disadvantages such as low efficiency, secondary pollution, and high energy consumption^[104]. Photocatalytic technology is considered as an effective ways to solve organics pollution due to its high efficiency, cleanliness, and sustainability^[105]. 2D carbon based photocatalysts, especially g-C₃N₄, MXene, and graphene, which have suitable band structures and

ideal light response, have become research hotspots in toxic organics degradation field.

Typically, Wang *et al.* developed an edge-functionalized g-C₃N₄ nanosheet photocatalyst [Figure 12a and b]^[106], which achieved efficient organic pollutant degradation by precisely controlling the edge structure of C₃N₄. The molecular engineering strategy was adopted to introduce functional groups such as amino and carboxyl groups at the edges of g-C₃N₄ nanosheets, significantly enhancing the photocatalytic activity. Experimental results show that the edge-functionalized nanosheets not only enhances the adsorption capacity of pollutants, but also optimizes the charge separation efficiency, resulting in a 5-fold increase in photocatalytic phenolic pollutant degradation rate compared to the pristine g-C₃N₄. In practical water treatment applications, the edge-functionalized g-C₃N₄ nanosheets have excellent degradation ability (removal rate over 95%) for various organic pollutants such as phenolic compounds and drug residues, thus endowing great application value in solving drinking water safety issues [Figure 12c]. Similarly, the porous g-C₃N₄ nanosheets [Figure 12d] with excellent visible light catalytic activity were designed^[107]. By optimizing the pyrolysis process, the rich pore structure and increased specific surface area of g-C₃N₄ nanosheets were successfully achieved. The characterization results indicate that the porous structure not only provides more active sites, but also significantly improves the light absorption performance and charge separation efficiency [Figure 12e]. Under visible light irradiation, the porous g-C₃N₄ nanosheets exhibits efficient degradation efficiency of organic methyl orange pollutants, being 4 times higher compared to traditional bulk g-C₃N₄ [Figure 12f]. Mechanism studies demonstrated that porous structures promote the generation of reactive oxygen species ($\cdot\text{OH}$ and $\cdot\text{O}_2^-$), which is a key factor in achieving efficient organic pollutant decomposition. In addition, the porous g-C₃N₄ nanosheets also exhibit good cycling stability, maintaining over 90% photocatalytic degradation activity after 5 cycles.

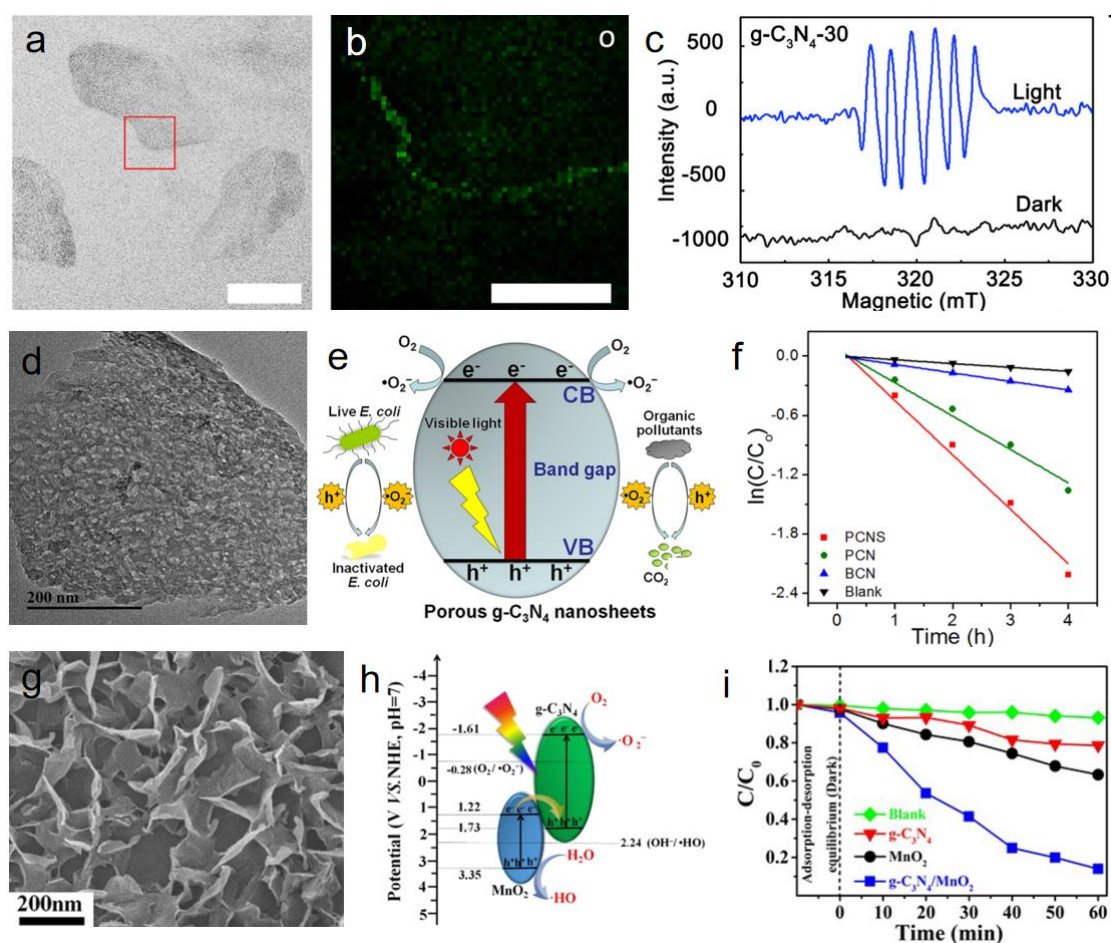


Figure 12. The (a) TEM, (b) SEM mapping of oxygen element, and (c) ESR spectra of the prepared edge-functionalized g-C₃N₄ nanosheet photocatalyst. Reproduced with permission.^[106] Copyright 2019, Elsevier; (d) TEM image, (e) schematic photocatalytic degradation, and (f) time dependent pollutant elimination over the porous g-C₃N₄ nanosheets. Reproduced with permission.^[107] Copyright 2017, American Chemical Society. (g) SEM image, (h) schematic photocatalytic degradation and electron shift, and (i) time dependent pollutant elimination over the 2D g-C₃N₄/MnO₂ composite photocatalyst. Reproduced with permission.^[108] Copyright 2018, American Chemical Society.

In addition to the modification of single 2D g-C₃N₄ catalyst systems, the 2D g-C₃N₄ based heterostructures for organic elimination have been also investigated. Yu's team reported the 2D/2D g-C₃N₄/MnO₂ nanocomposite photocatalyst [Figure 12g] with a Z-type charge migration route for efficient pollutant degradation [Figure 12h]^[108]. The

unique 2D/2D Z-type heterostructure was constructed by a simple hydrothermal method, which resulted in a tight interface contact between 2D g-C₃N₄ nanosheets and 2D MnO₂ nanosheets. It was revealed that this unique 2D structure not only provides a large number of active sites, but also establishes effective Z-type charge transfer channels. Moreover, the 2D g-C₃N₄/MnO₂ composite exhibits significantly enhanced photogenerated carrier separation efficiency and visible light absorption capability. Photocatalytic experiments demonstrate the 2D g-C₃N₄/MnO₂ composite exhibited 3.2 times higher photocatalytic Rhodamine B degradation than that of bulk g-C₃N₄ [Figure 12i], and had good cycling stability. Mechanism analysis illustrated that the Z-type electron transfer pathway effectively preserves electrons and holes with high oxidation-reduction ability, while suppressing carrier recombination.

Apart from 2D g-C₃N₄ materials, 2D graphene and MXene based materials have been also considered as efficient photocatalysts for environment remediation^[109-111]. The researchers constructed a 2D graphene oxide (GO) and MgFe₂O₄/ZnO heterojunction composite photocatalyst system by hydrothermal method^[112]. The introduction of GO in the ternary photocatalyst not only provides a larger specific surface area (about 180 m² g⁻¹), but also significantly enhances the electron transport capability, resulting in a 3.8 times increase in the efficiency of photogenerated carrier separation. Under visible light irradiation, the photocatalytic degradation efficiency of the 2D GO/MgFe₂O₄/ZnO heterojunction for methyl orange pollutants reached 98%, with a degradation rate constant 5.2 times higher than that of pure ZnO sample. Notably, the electron paramagnetic resonance (EPR) and capture experiments proved that the ternary photocatalyst system simultaneously generates abundant ·OH and ·O₂⁻ reactive species through Z-type charge transfer mechanism [Figure 13a], which exert significant function to achieving efficient pollutant degradation. In addition, the introduction of magnetic MgFe₂O₄ component enables the photocatalyst to be easily recovered through an external magnetic field, and maintains over 80% photocatalytic activity [Figure 13b] after 5 cycles, which provides a new approach for designing efficient and recyclable multi-component photocatalysts.

In another work, the researchers constructed a 2D V₂C MXene@g-C₃N₄ nanosheets heterojunction composite by a one pot method [Figure 13c]^[113]. By precisely controlling the synthesis conditions, a tight interface contact between V₂C MXene and g-C₃N₄ was achieved, forming a unique 2D/2D heterostructure. This 2D composite material exhibits the ultra-high specific surface area (about 320 m² g⁻¹) and rich surface functional groups from V₂C MXene, which endow excellent pollutant adsorption capacity. Moreover, the metallic conductivity of V₂C MXene leads to extraordinary photogenerated carrier separation. The *in-situ* XPS and DFT calculations confirmed that the Ti-N bond formed at the V₂C MXene@g-C₃N₄ interface promotes the transfer of electrons from g-C₃N₄ to V₂C MXene, establishing an efficient Z-type charge transfer channel [Figure 13d]. The photocatalytic experimental results suggested that the 2D V₂C MXene@g-C₃N₄ composite achieved a methylene blue removal efficiency of 99.5% under visible light irradiation, with adsorption contributing about 35% and photocatalytic degradation contributing about 64.5%.

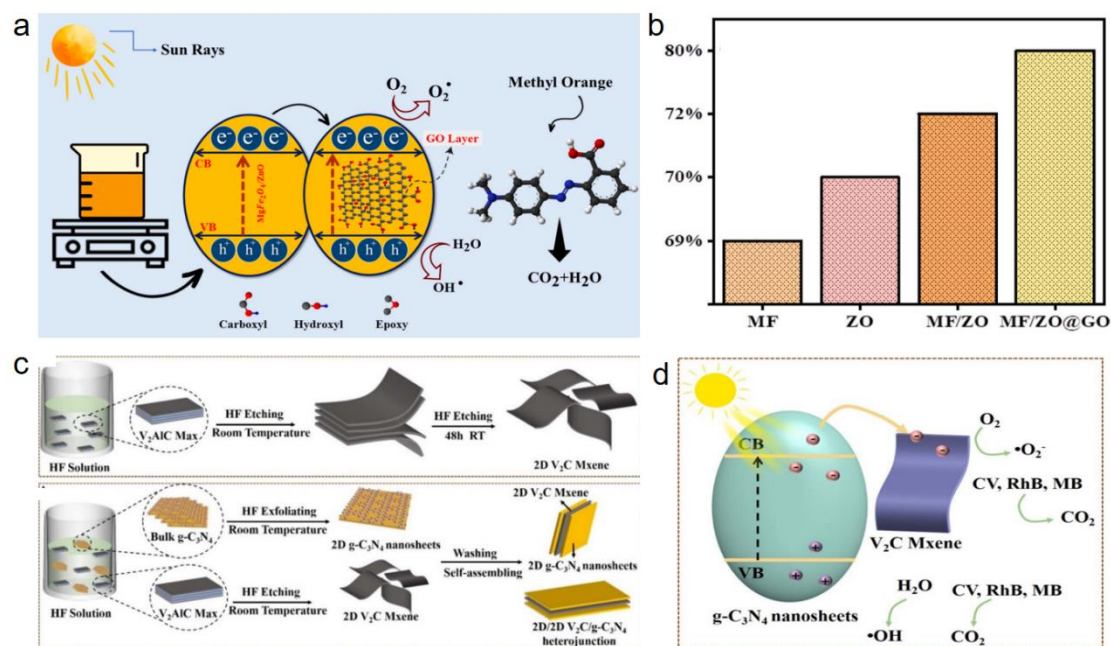


Figure 13. (a) The schematic mechanism photocatalytic pollution degradation and (b) degradation efficiency over the 2D GO/MgFe₂O₄/ZnO heterojunction. Reproduced

with permission.^[112] Copyright 2023, Elsevier; (c) The schematic synthesis process of the 2D V₂C MXene@g-C₃N₄ nanosheets heterojunction and (d) the photocatalytic organic pollutant degradation mechanism of the photocatalyst. Reproduced with permission.^[113] Copyright 2025, Royal Society of Chemistry.

Although 2D carbon based photocatalysts exhibit significant performance in the degradation of organic pollutants, it still faces problems such as low visible-light utilization, poor anti-interference ability in complex water bodies, and insufficient long-term stability. It is necessary to accurately design the electronic structure of catalysts to broaden the light response range, combine in-situ characterization to reveal degradation mechanisms, develop efficient recycling and immobilization technologies, and promote their large-scale application in the treatment of organic pollution in actual water bodies.

CONCLUSION AND OUTLOOKS

In this work, the common 2D carbon based materials (such as graphene, MXene, g-C₃N₄ etc.) for photocatalytic energy conversion and environment remediation including water splitting, CO₂ reduction, H₂O₂ generation, and pollutant degradation. Due to their unique 2D layered structure, high specific surface area, and excellent optoelectronic properties, the 2D carbon based photocatalyst materials can capture visible light and produce active substances (such as $\cdot\text{OH}$, $\cdot\text{O}_2^-$), thereby efficiently accelerating photocatalytic reactions. The common modification strategies including element doping, constructing heterojunctions, defect engineering, etc. for further increased photocatalytic performance were also elucidated. This critical review can deliver the better knowledge of the preparation of advanced photocatalyst and inspire innovative concepts for constructing efficient photocatalysis system. However, from basic research to practical applications of 2D carbon based photocatalysts, a series of challenges and inherent shortcomings were also presented, which were mainly discussed in the following four aspects:

1. The cost of large-scale preparation and commercialization

At present, the preparation of high-quality 2D carbon materials, such as few layer graphene and defect controllable g-C₃N₄, usually relies on high temperature and high pressure such as calcination and chemical vapor deposition. These methods have high energy consumption, low yield, and are prone to introducing uncontrollable defects, resulting in significant performance differences between batches, which cannot meet the requirements of stable supply chains and cost control for industrial applications. Thus, developing green and low-cost synthesis pathways is necessary. For example, exploring synthesis technologies based on biomass waste (such as cellulose and lignin) or inexpensive precursors is promising.

2. Insufficient long-term stability in practical application

Laboratory research often evaluates activity of photocatalysts within hours, but commercialization requires materials to operate stably for months or even years. In actual operation, photocatalysts face the problem of rapid performance degradation caused by multiple factors such as photocorrosion, chemical poisoning, mechanical wear, and biological pollution. Therefore, material structure strengthening should be considered. By crystallizing the material, coating it with a carbon layer, or constructing a core-shell structure, its chemical and photochemical inertness could be enhanced to resist corrosion and poisoning.

3. Recycling powdered photocatalysts

The long-standing problem of recycling powdered catalysts is particularly prominent in large-scale applications. Simple suspended reactors have high efficiency but huge separation costs. However, the immobilization technologies such as coating and embedding often lead to the shielding of photocatalyst active sites and an increase in mass transfer resistance, resulting in much lower reactor efficiency than laboratory

beaker experiments, and the energy consumption and cost per unit processing capacity are not competitive. Thus, developing in-situ growth technology such as directly growing 2D materials vertically on carriers with large specific surface area is a promising approach.

4. Weak anti-interference ability in complex water environments

Photocatalytic reactions are usually conducted upon water environment. However, the actual composition of water bodies is complex, which may contain various organic compounds, anions suspended solids, etc. They compete to absorb photons, quench active species, and occupy active sites on the catalyst surface, resulting in significantly decreased catalytic efficiency in practical applications. Thus, the enhancement of the specificity and stability of the 2D carbon catalytic system should be conducted in the future research.

In summary, in order to truly achieve industrial application of 2D carbon based photocatalysts, it is necessary to focus on tackling the core challenges of material stability and preparation based on an understanding of their basic physical and chemical processes. Nevertheless, 2D carbon based photocatalytic system is still considered as highly promising direction to address energy and environmental problems.

DECLARATIONS

Authors' contributions

Writing-original draft: Zhu, C.; Feng, A.; Li, Z.; Zhang, S.

Investigation: Ding, Y.; Li, H.; Zheng, R.; Wang, C.

Data curation, validation: Ding, Y.; Chen, L. H.; Su, B. L.

Conceptualization, methodology, resources: Ding, Y.; Wang, C.; Su, B. L.; Chen, L. H.

Availability of data and materials

Not applicable.

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Conflicts of interest

Su, B. L. is Editor-in-Chief of the journal *Chemical Synthesis*. Su, B. L. was not involved in any steps of editorial processing, notably including reviewers' selection, manuscript handling and decision making. The other authors declare that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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