

Applications of g-C₃N₄-Based Photocatalysts

Subjects: [Chemistry](#), [Physical](#)

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The assembly of g-C₃N₄ with metal oxides is an effective strategy which can not only improve electron–hole separation efficiency by forming a polymer–inorganic heterojunction, but also compensate for the redox capabilities of g-C₃N₄ owing to the varied oxidation states of metal ions, enhancing its photocatalytic performance. Applications of g-C₃N₄-based materials in photocatalysis are discussed, including water splitting to generate H₂ and O₂, the degradation of pollutants, CO₂ reduction and bacterial disinfection.

g-C₃N₄

water

synthesis

1. Photocatalytic Water Splitting for H₂

Because of the decreasing storage of fossil fuels and their negative impacts on the environment (releasing CO₂ for example), the use of green and renewable hydrogen fuels attracts much attention from scientists. The photocatalytic splitting of water is an ideal way to generate hydrogen and has become a hot topic in recent years. **Figure 1** presents a simplified diagram of splitting water into hydrogen and oxygen over g-C₃N₄ under light irradiation. First, g-C₃N₄ is excited by photons to generate electrons, which then jump to the CB, leaving holes at the VB. The photogenerated e⁻ and h⁺ flow to the surface of g-C₃N₄, reducing and oxidizing the adsorbed water to hydrogen and oxygen, respectively. However, the generated e⁻/h⁺ will rapidly recombine each other due to the Coulombic attraction, losing activity. The improvement in the separation efficiency of the photogenerated e⁻/h⁺ pairs, thus, is a challenging topic in the field of g-C₃N₄ photocatalysis.

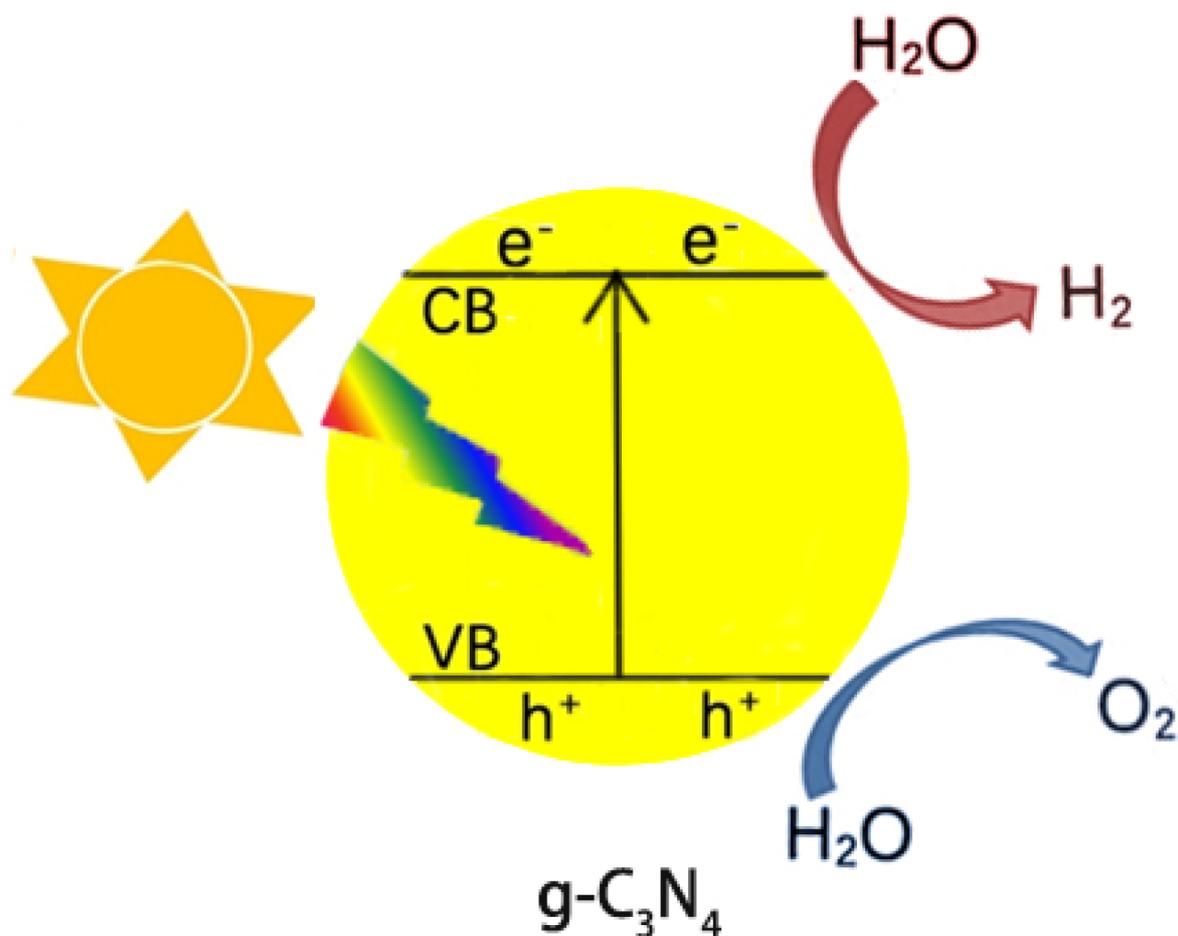


Figure 1. Scheme of photocatalytic water splitting into H₂ and O₂ over g-C₃N₄ under light irradiation.

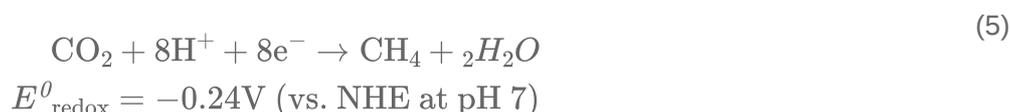
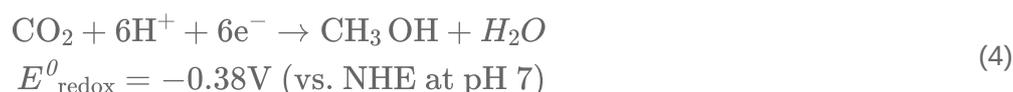
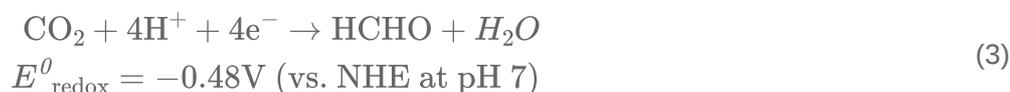
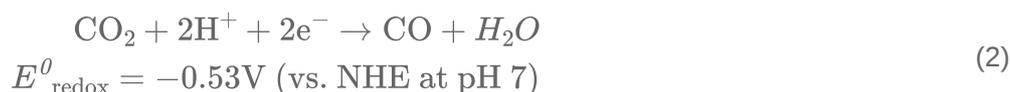
To achieve this, the coupling of g-C₃N₄ with metal oxide is a solution, which can separate e⁻/h⁺ pairs in space by forming an opposite flow of e⁻ and h⁺ (for type II heterojunctions), or by inducing the recombination of unused e⁻ and h⁺ (for Z-Scheme heterojunctions), as reported in the literature [1][2]. Shi et al. [3] reported the in situ synthesis of MoO₃/g-C₃N₄, via co-pyrolysis of MoS₂ and melamine, for photocatalytic water splitting to hydrogen, finding that the activity of g-C₃N₄ was significantly enhanced with the increase in MoO₃ content. It is possible that the use of layered MoS₂ as a precursor not only improves the dispersion of MoO₃ on g-C₃N₄, but also enhances the interactions between them. Li et al. [4] synthesized W₁₈O₄₉/g-C₃N₄ composites by roasting a g-C₃N₄-impregnated ammonium tungstate solution. The loading of W₁₈O₄₉ greatly improves the surface area (by about five times) and exhibits excellent activity for a photocatalytic hydrogen evolution reaction, with a reaction rate of 912.3 μmol·g⁻¹·h⁻¹, which is 9.7 times higher than that of g-C₃N₄.

The coupling of g-C₃N₄ with two metal oxides could be more interesting when compared to that with single-metal oxide, as multiple heterojunctions can be established, exhibiting rich optical properties, and hence, better photocatalytic activities. This is observed in many studies [5][6][7]. For example, Wang et al. [8] found that Fe₂O₃@MnO₂ core-shell g-C₃N₄ ternary composites can form double heterojunctions, which provide abundant channels for electrons transfer, exhibit enhanced optical properties and allow the two half-reactions (the production

of hydrogen and oxygen) to occur on the opposite surfaces of the semiconductor ; this results in improved activity for both hydrogen and oxygen production, with an optimal reaction rate of 124 $\mu\text{mol}\cdot\text{h}^{-1}$ and 60 $\mu\text{mol}\cdot\text{h}^{-1}$, respectively.

2. Photocatalytic Reduction of CO₂ to Renewable Hydrocarbon Fuels

With increasing global warming, it is critical to find effective ways to deal with greenhouse gases. Carbon dioxide (CO₂) is not only a typical greenhouse gas but also a valuable C1 resource. Hence, utilizing solar energy to reduce CO₂ into higher-value chemicals shows great advantages in solving the problems of both global warming and energy crises. In the past few years, g-C₃N₄ has been employed as a photocatalyst for CO₂ reduction owing to its high CB potential, which can activate CO₂ by donating electrons to the unoccupied orbits of CO₂. The photocatalytic CO₂ reduction involves a proton-assisted multi-electron process, as shown in Equations (1)–(5) below [9]. From the viewpoint of thermodynamics, CO₂ is gradually reduced to HCOOH, CO, HCHO, CH₃OH and CH₄ by receiving multiple (2, 2, 4, 6 and 8) electrons and protons, accompanying the increase in reduction potential. This means that the photocatalyst used to reduce CO₂ should have strong redox capability in order to supply sufficient driving force for the reaction.



ZnO can absorb CO₂ and has a CB potential (E_{CB}) of -0.44 eV, which is more negative than the reduction potential of CO₂. Therefore, the combination of ZnO and g-C₃N₄ would benefit the CO₂ reduction reaction. Indeed, it is found that although the deposition of ZnO has negligible effects on the light absorption capacity and surface area of g-C₃N₄, the ZnO/g-C₃N₄ composite shows better photocatalytic activity for CO₂ reduction than individual ZnO and g-C₃N₄, due to the formation of heterojunctions that facilitate the separation of e⁻/h⁺ pairs [10]. The CO₂ conversion rate obtained from ZnO/g-C₃N₄ reaches 45.6 μmol/g/h, which is 4.9 times and 6.4 times higher than that obtained from g-C₃N₄ and P25, respectively. Additionally, based on the fact that the zeta potential of ZnO is positive and that of g-C₃N₄ is negative, Nie et al. [11] constructed a ZnO/g-C₃N₄ composite using an electrostatic self-assembly method. The combination of them induces synergistic effects that are conducive to photocatalytic reactions, in which the ZnO microsphere prevents falling g-C₃N₄ nano flakes from gathering, and the g-C₃N₄ improves light utilization efficiency through the multi-scattering effect.

In addition to ZnO, many other metal oxides can couple with g-C₃N₄ and contribute to the CO₂ reduction reaction. For example, Bhosale et al. [12] employed a wet chemical method to couple FeWO₄ with g-C₃N₄, forming a Z-scheme g-C₃N₄/FeWO₄ photocatalyst; it showed good activity for the reduction of CO₂ to CO without any medium, with a CO production rate of 6 μmol/g/h, which is 6 and 15 times higher than that of individual g-C₃N₄ and FeWO₄.

3. Photocatalytic Degradation of Pollutants

With the rapid development of the economy, various toxic pollutants emitted from industrial plants have been discharged to the environment and have seriously destroyed the ecological system. The removal of pollutants and the remediation of the environment have thus become essential topics and have attracted broad attention in recent years. Photocatalysis is a prospective technology for pollutant removal, and is able to mineralize organic pollutants into CO₂ and H₂O by producing oxidizing intermediates (such as •O₂⁻, •OH and h⁺). Depending on the properties of the pollutants, three reaction types can be classified: (1) the removal of organic pollutants in aqueous solution, such as dye [4][13] and antibiotic degradation [14]; (2) the removal of heavy-metal cations in aqueous solution, such as the reduction of chromium (VI) [15]; and (3) the removal of organic or inorganic pollutants in gas phase, such as the degradation of ortho-dichlorobenzene [16], acetaldehyde [17] and nitric oxide [18].

The Fenton advanced oxidation process (with an Fe²⁺ and H₂O₂ system) is a traditional technology used to treat industrial wastewater, but it is limited to a narrow pH range (<3) and causes secondary pollution due to the production of iron sludge. For this reason, it is proposed that a photocatalyst should be used instead of Fe²⁺, to activate H₂O₂ into •OH radicals under light irradiation conditions, which can be achieved in a wide pH range without producing secondary pollutants. Hence, it is a green route to removing organic pollutants in aqueous solution and has good prospects for industrial use.

In this respect, Xu et al. [19] recently reported that the LFO@CN photocatalyst is highly efficient for the oxidative degradation of RhB with H₂O₂ under visible-light irradiation, with 98% conversion obtained within 25 min, and the material can be recycled for four cycles with no appreciable deactivation. Moreover, when applying a ternary LaFe_{0.5}Co_{0.5}O₃/Ag/g-C₃N₄ heterojunction that consists of a redox part LaFe_{0.5}Co_{0.5}O₃ (LFCO), photo part g-C₃N₄ and plasmonic part (Ag), for the degradation of tetracycline hydrochloride (TC), in the presence of H₂O₂ and light irradiation, the system exhibits good activity due to a photo-Fenton effect induced in the reaction. In this system, H₂O₂ is first activated into •OH radicals and OH⁻ anions over the LFCO, and the OH⁻ anions subsequently react with holes (h⁺) produced at the VB band of LFCO to form more •OH radicals. Hence, H₂O₂ can be fully utilized to oxidize TC in the reaction. Meanwhile, the O₂ dissolved in the solution can react with the electrons (e⁻) generated at the CB band of g-C₃N₄ and form •O₂⁻, which is also a strong oxidant that is able to oxidize TC into CO₂ and H₂O. These results support that g-C₃N₄-based catalysts have good chemical stability and can be an effective substitute for Fenton catalysts in environmental purification.

In addition to the direct addition of H₂O₂, the photocatalytic in situ generation of H₂O₂ in the reaction for pollutant oxidation, which is a more promising way but a more challenging topic, is also possible. For example, Xu et al. reported that ternary g-C₃N₄/Co₃O₄/Ag₂O heterojunctions can accelerate the mineralization of RhB due to the presence of H₂O₂ in situ, produced from O₂ reduction [20]. Through studying the catalytic behavior of the composites in the electrochemical oxygen reduction reaction (ORR), they found that the average number of electrons transferred in the reaction is 2.07, which indicates that the two-electron O₂ reduction process is the dominant step in the reaction.

The morphology of metal oxide, the interface interaction between metal oxide and g-C₃N₄ and the method of coupling metal oxide with g-C₃N₄ are also crucial factors affecting the photocatalytic performance of g-C₃N₄ for pollutant removal. For instance, the coupling of cubic CeO₂ (3–10 nm) with g-C₃N₄ using a hydrothermal method can greatly improve the activity of g-C₃N₄ for methyl orange degradation, with the reaction rate reaching 1.27 min⁻¹, which is 7.8 times higher than that of g-C₃N₄ alone (0.16 min⁻¹) [21]. The hybridization of NiO with g-C₃N₄ causes a red shift in the UV absorption edge and boosts the ability of light response; hence, it exhibits improved activity for methylene blue degradation, which is about 2.3 times higher than that of g-C₃N₄ [22]. Similar phenomena are also observed for other materials, e.g., TiO₂-In₂O₃@g-C₃N₄ [23].

The heavy-metal ions produced in electroplating, metallurgy, printing and dyeing, medicine and other industries cause serious damage to the ecological environment. Cr(VI) is a typical heavy metal in wastewater and its removal receives wide attention. The photocatalytic reduction of Cr(VI) to Cr(III) is an efficient way to treat Cr(VI)-containing wastewater, due to its simple process, energy savings, high efficiency and lower levels of secondary pollution [24]. It has been reported that the in situ self-assembly of g-C₃N₄/WO₃ in different organic acid media can lead to various surface morphologies and catalytic activities for Cr(VI) removal, as the number of carboxyl groups in organic acid greatly affects the shape and performance of g-C₃N₄/WO₃. Its synthesis in ethanedioic acid medium, which contains two carboxyl groups, yields a disc shape and has the best activity for nitroaromatic reduction. Furthermore, the material has good stability for the reaction, with no appreciable activity loss within four cycles.

Bi₂WO₆ is a promising semiconductor that can couple with g-C₃N₄ and form a heterojunction for the photocatalytic treatment of Cr(VI)-containing wastewater. Song et al. [25] found that a C₃N₄/Bi₂WO₆ composite prepared using a hydrothermal method exhibits a surface area up to 46.3 m²/g and shows a rate constant of 0.0414 min⁻¹ for the photocatalytic reduction of Cr(VI), as the high surface area of the catalyst facilitates not only the reactant's adsorption, but also the visible-light absorption.

Photocatalysis is also effective for removing gas-phase pollutants and receives great interest from scientists. It is known that air pollution is a big problem for the environment, and causes serious harm to the human body and ecological systems by forming acid rain, chemical smog, particulate matter, etc. Hence, seeking an effective and feasible technology for its removal is a challenging topic. Photocatalysis provides a way to remove air pollutants (e.g., NO_x) by installing catalysts either inside the exhaust pipe or on the road surface [26]. As a typical photocatalyst, g-C₃N₄-based materials are also widely investigated in this aspect. Zhu et al. reported that g-C₃N₄ is active in NO removal via thermal catalysis, and proposed that the N atoms of g-C₃N₄, with a lone electron pair, serve as the active site of NO by donating electrons to weaken the N-O bond order [27]. This lays the foundation for using photocatalysis for NO removal, as electrons can be effectively excited from g-C₃N₄ under light irradiation.

However, it is known that the surface area of g-C₃N₄ prepared using the thermal condensation method is small, which greatly limits the light absorption capacity, the e⁻/h⁺ separation efficiency and other physicochemical properties; thus, many strategies have been adopted to overcome this problem. For example, Sano et al. [28] reported that pretreating melamine with NaOH solution before the condensation process favors the hydrolysis of unstable domains and the generation of mesopores in the structure of g-C₃N₄, leading to an increase in surface area from 7.7 m²/g to 65 m²/g, and the NO oxidation activity is accordingly increased 8.6 times. Duan et al. [18] found that flower-like g-C₃N₄ prepared using the self-assembly method can notably improve photocatalytic activity for NO oxidation compared to bulk g-C₃N₄, owing to the enlargement of the BET surface area, the formation of nitrogen vacancies, the condensation of π-π layer stacking, and the improvement in e⁻/h⁺ separation efficiency. The alternation of the precursor, e.g., urea [29] and guanidine hydrochloride [30] is also efficient in preparing g-C₃N₄ with a large surface area and improving photocatalytic performance.

4. Sterilization and Disinfection

In addition to the above applications, photocatalysis is also widely applied to inactivate pathogens in surface water owing to its broad compatibility, long durability, anti-drug resistance and thorough sterilization [31]. Bacteria, such as salmonella, staphylococcus aureus and bacillus anthracis, are commonly used as model pathogens to evaluate photocatalytic disinfection efficiency. Since the first work of Matsunaga et al. [32] on photochemical sterilization in 1985, this technique has rapidly developed and receives great interest from scientists. The principle of photocatalytic sterilization is to excite and separate the e⁻/h⁺ pairs via illumination; the photoinduced electrons and/or holes then inactivate the bacteria by directly or indirectly inflicting oxidative damage on their organs (through the formation of •O₂⁻, •OH, etc.). Hence, the disinfection efficiency of materials closely depends on the properties that influence the generation and separation of e⁻/h⁺ pairs, e.g., the surface area, the band gap and the surface morphology, as reported for other photocatalytic processes.

In the case of g-C₃N₄, Huang et al. [33] found that mesoporous g-C₃N₄ synthesized using the hard template method can inactivate most of the bacteria (e.g., *E. coli* K-12) within 4 h, owing to its large surface area, which allows more active sites exposed on the surface to produce h⁺ for bacterial disinfection. To support that the inactivation of bacteria is caused by photocatalysis, Xu et al. [34] conducted a dark contrasting experiment using a porous g-C₃N₄ nanosheet (PCNS) as the photocatalyst and *E. coli* as the model bacteria; they found that the adsorption of *E. coli* on PCNS reaches equilibrium within 1 h and about 85.5% of *E. coli* survive after 4 h, while nearly 100% of *E. coli* are killed by PCNS within 4 h under visible-light irradiation. This demonstrates that the PCNS has little toxic effect on *E. coli* and the disinfection is mainly caused by the electrons or holes induced from PCNS under light irradiation.

In addition to bacterial infection, viral outbreaks, including SARS, bird flu, Ebola and the recent COVID-19, are also important events related to human health, and they are generally more resistant than bacteria to conventional disinfection due to their small size. Thus, the inactivation of viruses normally requires strong oxidative agents. g-C₃N₄-based materials have good photocatalytic reactivity to produce strong oxidative agents, e.g., •O₂⁻ and •OH; hence, they are potential photocatalysts for virus inactivation. It has been reported that phage MS₂ can be completely inactivated by g-C₃N₄ under visible-light irradiation within 360 min [35], and the main active species for the reaction are •O₂⁻ and •OH. The loss of protein triggers the leakage and rapid destruction of internal components, and ultimately leads to the death of the virus without regrowth.

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