

Photocatalytic Application in Energy and Environmental Sustainability

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The energy and environmental crises have been an ongoing challenge, which is related to the vital interests of people around the globe. How to solve this problem through sustainable development strategies is considered deeply by scientific researchers. Photocatalysis provides a powerful technique for fully utilizing solar in the field of energy conversion.

g-C₃N₄

preparation

modification

hydrogen evolution

CO₂ conversion

1. H₂ Production

Hydrogen is gathering strong momentum as a pivotal energy transition pillar driven by the global shift toward decarbonization. Nevertheless, 85% of H₂ is produced from fossil fuel combustion, which generates roughly 500 metric tons of carbon dioxide every year and proffers a challenge and obstacle toward the sustainable living of future generations ^[1]. Solar-driven photocatalytic H₂ generation as a promising technology has received extensive attention in addressing the global energy crisis ^{[2][3]}. Photocatalytic water splitting for the energy transformation from solar to eco-friendly fuels has been studied for decades with various semiconductor photocatalysts. As a type of semiconductor photocatalyst, g-C₃N₄ is simple and inexpensive to fabricate, and has an adequate bandgap (≈ 2.7 eV) for activation upon sunlight irradiation. Wang's group first utilized g-C₃N₄ in photocatalytic H₂ evolution ^[4] ^[5]. Nonetheless, pristine g-C₃N₄ is far from satisfactory energy conversion because of its low light energy utilization, low density active sites, and ineffective isolation of the photogenerated excitons. Thus, researchers have proposed numerous strategies to boost the photocatalytic activity of g-C₃N₄-based materials for H₂ production. For example, the g-C₃N₄/carbon-dot-based nanocomposites, which possess enormous visible light absorption and applicable energy structures, have been prepared and serve as efficacious photocatalysts in photocatalytic water splitting for H₂ generation under light illumination ^{[3][6][7][8]}. Gao et al. reported hexagonal tubular g-C₃N₄/CD-based nanocomposites which exhibited nine times higher than bulk g-C₃N₄ in H₂ production rate ^[3] and related results indicated that CDs performed as both photosensitizer and electron acceptor. CDs could absorb long wavelength light to extend the visible-light response region and suppress the recombination of electron-hole pairs. Hussien et al. ^[9] combined four different strategies (non-metal doping, porosity generation, functionalization with amino groups, and thermal oxidation etching) in a one-pot thermal reaction and successfully prepared amino-functionalized ultrathin nanoporous B-doped g-C₃N₄ by using NH₄Cl as a gas bubble template, together with a thermal exfoliation process to produce ultrathin sheets. According to the process, the surface area, adsorption

capacity, and charge migration of the as-prepared photocatalyst have been improved, and a $3800 \mu\text{mol g}^{-1} \text{h}^{-1}$ H_2 generation rate and 10.6% prominent quantum yield were recorded. Li et al. [10] decorated carbon self-doping $\text{g-C}_3\text{N}_4$ nanosheets with gold-platinum (AuPt) nanocrystals through a photo-deposition route and compared the photocatalytic H_2 evolution performance of Pt/CCN, Au/CCN, Au/Pt/CCN, and Pt/Au/CCN, in which AuPt/CCN stood out and gave the highest H_2 generation rate ($1135 \mu\text{mol h}^{-1}$). The excellent performance can be ascribed to the non-plasmon-related synergistic effect of Au and Pt atoms in AuPt nanocrystals. Sun et al. [11] assessed the arrangements of metal- and non-metal-modified $\text{g-C}_3\text{N}_4$ composites in hydrogen evolution and found that the contribution of dye conjugation in non-metallic $\text{g-C}_3\text{N}_4$ composites favored their performance. However, the co-catalyst doping strategy was recommended for metallic $\text{g-C}_3\text{N}_4$ composites. In addition, the hybrid of MOF materials and $\text{g-C}_3\text{N}_4$ is also a good approach to develop novel photocatalysts. For example, Devarayapalli et al. [12] reported a $\text{g-C}_3\text{N}_4/\text{ZIF-67}$ nanocomposite and obtained a $2084 \mu\text{mol g}^{-1} \text{H}_2$ production, which is 3.84-fold greater than that of bare $\text{g-C}_3\text{N}_4$.

2. CO_2 Photoreduction over $\text{g-C}_3\text{N}_4$

Rising atmospheric levels of CO_2 and the consumption of fossil fuels raise a concern about the continued reliance on the utilization of fossil fuels for both energy and chemical production [13]. Photocatalytic reduction of CO_2 is a promising strategy to meet increasing energy needs and reduce the greenhouse effect [14]. Through photocatalytic reduction, CO_2 can be converted to light oxygenates and hydrocarbons. Photocatalytic CO_2 reduction is a multielectron transfer process. Fu et al. [15] have listed the possible reaction and corresponding redox potentials and stated that CO_2 was complicated to reduce at room temperature due to its stable chemical structure. For the complex reaction, five factors, comprising the matching of band energy, separation of charge carrier, kinetic of e- and hole transfer to CO_2 and reductant, the basicity of photocatalyst, and the strength and coverage of CO_2 adsorption, are considered to be crucial [16]. As a hot member of photocatalysts, $\text{g-C}_3\text{N}_4$ has been applied to CO_2 photo-reduction in recent years because the CB of $\text{g-C}_3\text{N}_4$ is sufficient to reduce CO_2 to various hydrocarbons, such as CH_3OH , CH_4 , HCHO , and HCOOH and so on. [17].

However, metal-free $\text{g-C}_3\text{N}_4$ is limited for CO_2 reduction activity due to its poor ability to activate the C-O bond of CO_2 . To improve the photocatalytic movement of CO_2 conversion, different metal units have been composited with $\text{g-C}_3\text{N}_4$ for broadening the absorption response range, and accelerating the charge separation and transfer, such as Pt/ $\text{g-C}_3\text{N}_4$ [18], $\text{Co}^{2+}/\text{g-C}_3\text{N}_4$ [19][20], Au/ $\text{g-C}_3\text{N}_4$ [21] and so on. Metal nanoparticles acting as cocatalysts could effectively improve the photocatalytic activity and selectivity of CO_2 reduction. In addition, other methods, including doping, loading cocatalysts and nanocarbons, constructing Z-scheme, and heterojunction, have also been employed [22][23][24][25][26][27][28][29][30]. For example, Fu et al. [31] prepared hierarchical porous O-doped $\text{g-C}_3\text{N}_4$ nanotubes (OCN-Tube) through continuing thermal oxidation exfoliation and curling condensation of bulk $\text{g-C}_3\text{N}_4$. Due to the higher specific surface area, better light harvesting, higher CO_2 uptake capacity, and superior separation efficiency of photogenerated charge carriers, the OCN-Tube exhibits excellent photocatalytic CO_2 reduction performance into CH_3OH . The CH_3OH evolution rate was as high as $0.88 \mu\text{mol g}^{-1} \text{h}^{-1}$, five times higher than the bulk ($0.17 \mu\text{mol g}^{-1} \text{h}^{-1}$). Huo et al. [32] fabricated amine-modified step-scheme (S-scheme) porous $\text{g-C}_3\text{N}_4$.

C₃N₄/CdSe-diethylenetriamine (A-PCN/CdSe-DETA) by a one-step microwave hydrothermal method. The modification by amine and formation of S-scheme heterojunction contributed to the remarkable photocatalytic performance of A-PCN/CdSe-DETA composite in CO₂ reduction and a CO production rate of 25.87 μmol/(h g) was achieved under visible-light irradiation. Wang et al. [14] reviewed different modification methods of g-C₃N₄-based photocatalysts for CO₂ reduction. They discussed each method (including morphology adjustment, co-catalysts, heterostructures, and doping) and compared the theoretical calculations and experimental results. By morphology adjustment, g-C₃N₄ with various shapes can be fabricated, such as rods, tubes, nanosheets, hollow spheres, and honeycomb-like structures. Due to the advantage of cocatalysts (e.g., Au, Ag, Pt, Pd, MXene, AuCu alloy, Pd-Ag), g-C₃N₄ with co-catalysts can be widely applied to activate CO₂ on the surface. Heterojunction with different types is also an effective method to improve the properties of g-C₃N₄-based materials. In addition, elemental doping is considered a common method to enhance photocatalytic quantum efficiency by changing the energy band, surface electronic property, and electrical conductivity.

3. Degradation of Organic Pollutants

Along with rapid population growth and significant industrialization development, large numbers of toxic, hazardous, and endless contaminants invade the environment, threatening to human life, especially a variety of pollutants present in water that are difficult to eliminate or degrade naturally. Photocatalytic degradation of contaminants is a green and efficient technology for coping with sewage [6][33]. Different kinds of g-C₃N₄-based materials (**Table 1**) have been exploited to increase the photodecomposition efficiency of pollutants, such as the constructed heterojunction, loading O₂-reduction co-catalysts, g-C₃N₄/CDs-based nanocomposites, and so on [23][34][35][36]. Generally, under the irradiation of visible light, the photogenerated electrons (e⁻) on the g-C₃N₄ catalyst will be excited from VB to CB, leaving holes (h⁺) in the VB. The holes can oxidize pollutants directly or react with H₂O/OH⁻ to form hydroxyl radicals [37]. When the REDOX potential of g-C₃N₄ composites is more negative than O₂/O₂⁻, the photogenerated electrons in the material can react with O₂ to produce O₂⁻ with strong oxidation capacity [38]. In addition, the resulting O₂⁻ could be protonated to produce OH [39]. Finally, the RhB dye is degraded to CO₂ and H₂O under the action of these free radicals. Chen et al. [40] fabricated a BiFeO₃/g-C₃N₄ heterostructure through mixing-calcining and compared its performance with BiFeO₃. Around 30% higher photocatalytic efficiency toward RhB dye was observed for the BiFeO₃/10% g-C₃N₄ heterostructure, which was assigned to the contribution of a higher concentration of O₂⁻. Zhang et al. [41] studied the selective reduction of molecular oxygen on g-C₃N₄ and probed its effect on the photocatalytic phenol degradation process. Compared with bulk g-C₃N₄, the exfoliated nanosheet yielded a three times improvement in photocatalytic phenol degradation. It has been demonstrated that bulk g-C₃N₄ prefers to reduce O₂ to O₂⁻ via one-electron reduction. At the same time, the photoexcited g-C₃N₄ nanosheet facilitates the two-electron reduction of O₂ to yield H₂O₂ because of the formation of 1,4-endoperoxide species. The two-electron reduction of O₂ on the nanosheet surface boosts hole generation and thus accelerates phenol oxidation degradation [41][42]. Thus, to improve the photocatalytic performance of g-C₃N₄, more effort should be devoted to strengthening the solid O₂-reduction reactions. For example, Liu et al. [43] reported a heterojunction material of K-doped g-C₃N₄ nanosheet -CdS and degraded tetracycline with 94% degradation under visible light in 30 min. In addition, due to the electronegativities, ionic radius differences, and impurity states, element doping is

also an effective method to manipulate the electronic structure and physicochemical performance of g-C₃N₄-based materials. Gao et al. [44] synthesized Fe-doped g-C₃N₄ nanosheets and obtained 1.4- and 1.7-fold higher degradation rates of MB than that of pure g-C₃N₄ nanosheets and bulk g-C₃N₄, which indicated that the exploitation of efficient g-C₃N₄-based photocatalysts with high stabilization and degradation under visible light irradiation would significantly contribute to sewage disposal. Zhang et al. [45] synthesized a novel hybrid of Zr-based metal-organic framework with g-C₃N₄ (UiO-66/g-C₃N₄) nanosheets and applied a photodegradation of methylene blue, by which a 100% photodegradation was achieved within 4 h under visible light. Here, it has been provided with a new insight into the design of g-C₃N₄-based photocatalysts to deal with organic dyes in the environment.

Table 1. Photocatalytic degradation of pollutants over g-C₃N₄-based materials reported within the last three years.

Entry	Photocatalyst	Pollutant Concentration	Light Source	Degradation Efficiency/%	Ref.
1	5% g-C ₃ N ₄ -TiO ₂	Acetaminophen: 0.033 mM	300 W Xe (>400 nm)	99.3 in 30 min	[46]
2	3ZIF/1.5Au-PCN	Bisphenol A	350 W Xe (>420 nm)	>85%	[47]
3	Cu(tmpa)/20%CN	Congo red: 100 mg·L ⁻¹	150 W Xe	98.2% in 3 min	[48]
4	BiO-Ag(0)/C ₃ N ₄ @ZIF-67	Congo red: 12 mg·L ⁻¹	Natural sunlight	90% in 150 min	[49]
5	C ₃ N ₄ /RGO/Bi ₂ Fe ₄ O ₉	Congo red: 10 mg·L ⁻¹	LED 30 W	87.65% in 60 min	[50]
6	g-C ₃ N ₄ /Co-MOF	Crystal violet: 4 ppm	MaX 303 solar simulator (50 mW/cm ²)	95% in 80 min	[12]
7	Honeycomb-like g-C ₃ N ₄ /CeO ₂ -x	Cr (VI): 20 mg·L ⁻¹	300 W Xe (>420 nm)	98% in 150 min	[51]
8	Sm ₆ WO ₁₂ /g-C ₃ N ₄	Levofloxacin: 10 mg·L ⁻¹	150 Mw cm ⁻² tungsten lamp	98% in 70 min	[52]
9	O-g/C ₃ N ₄	Lincomycin: 100 mg·L ⁻¹	PCX50C system (>420 nm)	99% within 3 h	[53]
10	ZnO-modified g-C ₃ N ₄	Methylene blue: 10 ppm	200 W tungsten lamp (>420 nm)	97% in 80 min	[54]
11	Wood-like g-C ₃ N ₄ @WDC	Methylene blue: 20 mg·L ⁻¹	300 W Xe (>400 nm)	98% in 60 min	[55]

Entry	Photocatalyst	Pollutant Concentration	Light Source	Degradation Efficiency/%	Ref.
12	BiO-Ag(0)/C ₃ N ₄ @ZIF-67	Methylene blue: 12 mg·L ⁻¹	Natural sunlight	96.5% in 120 min	[49]
13	Cerium-based GO/g-C ₃ N ₄ /Fe ₂ O ₃	Methylene blue: 10 mg·L ⁻¹	Light bulb	70.61% in 45 min	[56]
14	Ytterbium oxide-based GO/g-C ₃ N ₄ /Fe ₂ O ₃	Methylene blue: 10 mg·L ⁻¹	Light bulb	83.5% in 45 min	[56]
15	Cu(tmpa)/20%CN	Methylene blue: 10 mg·L ⁻¹	150W Xe	92.0% within 20 min	[48]
16	C ₃ N ₄ _x /AgO _y @Co _{1-x} Bi _{1-y} O ₇	Methylene blue: 25 mL 10 mM	100 W tungsten bulb	96.4% in 120 min	[57]
17	Ternary composites of Zr-MOF combined with g-C ₃ N ₄ and Ag ₃ PO ₄	Methylene blue: 10 mg·L ⁻¹	85-watt tungsten lamp outdoor/solar light in an open air	95% within 240 min 93% within 105 min	[58]
18	PSCN/Ag@AgI/WO ₃	Malachite green: 1 × 10 ⁻⁴ mol dm ⁻³	35 W LED	90% in 60 min	[59]
19	Cu(tmpa)/20%CN	Malachite green: 30 mg·L ⁻¹	150W Xe	92.9% in 35 min	[48]
20	20% g-C ₃ N ₄ /Bi ₄ O ₅ I ₂	Methyl orange: 20 mg·L ⁻¹	350 W Xe	0.164 min ⁻¹	[60]
21	Cu(tmpa)/20%CN	Methyl violet: 10 mg·L ⁻¹	150W Xe	92.0% in 60 min	[48]
22	MnCo ₂ O ₄ /g-C ₃ N ₄	Nitrobenzene: 40 mg L ⁻¹	CMCN2/PMS system	96.7% in 240 min	[61]
23	C ₃ N ₄ _x /AgO _y @Co _{1-x} Bi _{1-y} O ₇	Oxytetracycline: 25 mL 25 mM	100 W tungsten bulb	93% in 160 min	[57]
24	g-C ₃ N ₄ /WO ₃ /WS ₂	Rhodamine B: 25 mg L ⁻¹	300 W Xe (>420 nm)	96.2% in 20 min	[62]
25	Flower-like Bi ₁₂ TiO ₂₀ /g-C ₃ N ₄	Rhodamine B: 20 mg·L ⁻¹	150 mW·cm ⁻² Xe (>420 nm)	100% in 30 min	[63]
26	CdS/CQDs/g-C ₃ N ₄	Rhodamine B: 10 mg·L ⁻¹	300 W Xe (>420 nm)	100% in 20 min	[64]

Entry	Photocatalyst	Pollutant Concentration	Light Source	Degradation Efficiency/%	Ref.
27	Ytterbium oxide-based GO/g-C ₃ N ₄ /Fe ₂ O ₃	Rhodamine B: 10 mg·L ⁻¹	Light bulb	67.11% in 45 min	[56]
28	Cerium-based GO/g-C ₃ N ₄ /Fe ₂ O ₃	Rhodamine B: 10 mg·L ⁻¹	Light bulb	63.08% in 45 min	[56]
29	Fish-scale g-C ₃ N ₄ /ZnIn ₂ S ₄	Tetracycline: 10 mg·L ⁻¹	300 W Xe (>420 nm)	74% in 30 min	[65]
31	Flower-like Co ₃ O ₄ /g-C ₃ N ₄	Tetracycline: 15 mg·L ⁻¹	350 W Xe (>420 nm)	85.32% in 120 min	[66]
31	10 wt% CuAl ₂ O ₄ /g-C ₃ N ₄	Tetracycline hydrochloride: 100 mg·L ⁻¹	300 W Xe (>400 nm)	89.6% in 60 min	[67]
32	CO-C ₃ N ₄	Tetracycline hydrochloride: 10 mg·L ⁻¹	300 W Xe (>420 nm)	97.77% (PMS) in 40 min	[68]
33	ZIF-67/g-C ₃ N ₄	Venlafaxine: 10 mg·L ⁻¹	-	27.75% within 120 min	[69]
34	ZIF-67/MIL-100(Fe)/g-C ₃ N ₄	Venlafaxine: 10 mg·L ⁻¹	-	100% within 120 min	[69]
35	ZIF-67/MOF-74(Ni)/g-C ₃ N ₄	Venlafaxine: 10 mg·L ⁻¹	-	91.8% within 120 min	[69]

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