

Advancing photocatalytic oxidation process for sustainable treatment of wastewater from livestock production: current breakthroughs and key challenges

Bo SUN¹, Xiaona PAN², Xingxing QIAO¹, Wenlong BI¹, Yichen HAO¹, Junmei QIN¹, Qingjie HOU (✉)¹, Fenwu LIU (✉)¹

1 College of Resource and Environment, Shanxi Agricultural University, Taigu 030801, China.

2 Department of Basic Sciences, Shanxi Agricultural University, Taigu 030801, China.

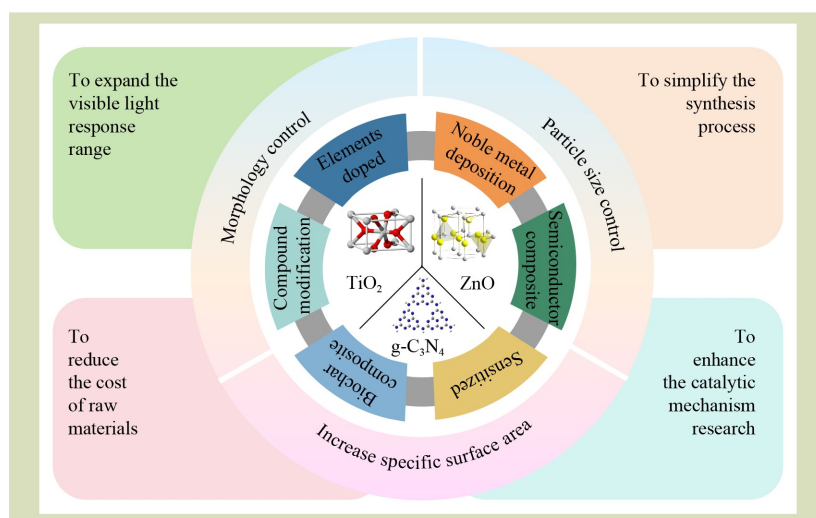
KEYWORDS

Cost-effectiveness, degradation pathways, modification methods, organic pollutants, visible-light-driven

HIGHLIGHTS

- Adjusting material morphology, particle size and SSA enhances catalytic activity.
- Organic pollutants are degraded by reactive free radicals produced by the catalyst.
- Optimizing energy and material use during synthesis is highlighted.
- Effective use of waste enables circular and low-cost photocatalyst modification.
- Potential approaches for cost control and catalytic applications are outlined.

GRAPHICAL ABSTRACT



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Correspondences: qingjiehou@163.com,
lffw2008@163.com

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ABSTRACT

Wastewater from livestock production is characterized by a complex composition, high pollutant load and the presence of emerging contaminants. These properties lead to critical challenges in conventional treatment processes, including excessive energy consumption, low treatment efficiency and incomplete pollutant removal. Photocatalytic oxidation is an advanced oxidation process that uses light energy to generate reactive oxygen species to degrade pollutants. It has gained significant attention due to its advantages of high efficiency, environmental friendliness and the ability to mineralize organic pollutants into water, carbon dioxide and other small molecules without consuming fossil energy. However, despite its potential, photocatalytic oxidation has not been widely applied in wastewater treatment. This is mainly

due to the large band gap, low utilization of visible light and fast carrier recombination of photocatalyst. To address these issues, this paper comprehensively reviews the current technical developments of the photocatalytic oxidation process and suggests potentially productive future studies. Despite significant progress, several critical challenges remain to be addressed in photocatalytic material applications, including low visible light utilization, complex synthesis process, expensive material costs, poor practical performance and insufficient mechanism understanding. This review will help design high-efficiency visible-light-driven photocatalysts and promote the application of photocatalysts in the treatment of wastewater from livestock production.

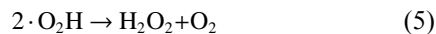
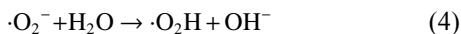
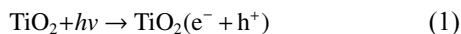
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1 Introduction

Large livestock industries discharge substantial amounts of wastewater with high concentration organic pollutants, such as residual veterinary drug antibiotics, antibiotics, pathogens and heavy metals, which are a serious threat to the ecological safety of the surrounding water bodies^[1-4]. The metabolites of antibiotics and livestock constantly produce resistant bacteria in the environment and stimulate the dosage of drugs^[5,6]. It caused a vicious circle of with drugs contaminating the environment leading to more drug-resistant bacteria that results in increased drug dosage being used and then increased environmental contamination. Due to their hard-degradation nature, these pollutants are inefficiently degraded by conventional processes, and both the pollutants and their degradation intermediates enter water bodies with waste water treatment plants effluent^[7]. Given that the vast majority of pollutants are water-soluble, these substances are eventually enriched in the water environment, and then enter the human body through the food chain, causing irreversible toxic operation^[8].

Using physical adsorption to remove the organic pollutants from wastewater from livestock production is inefficient^[9]. While physical adsorption can temporarily immobilize contaminants through surface interactions, it fundamentally lacks degradation capability, thereby posing risks of secondary pollution from desorption or residual pollutants^[10,11]. Also, biological treatment is susceptible to the effect of inhibitory compounds in the wastewater, which can reduce their processing effect^[12,13]. Due to physical adsorption and biological treatment processes cannot effectively remove the

pollutants in the aquaculture wastewater, the advanced oxidation processes rise in response to the proper time and conditions^[14]. These processes are a group of water treatment technologies that use highly reactive oxidizing species to degrade and remove persistent organic pollutants, transforming them into less harmful substances like carbon dioxide and water. These mainly include the Fenton, photocatalytic, ultrasonic and supercritical water processes. Compared with other methods, photocatalytic oxidation shows many advantages, such as simple reaction equipment and safe operation, low cost and high efficiency, and no secondary pollution^[15-17]. Since TiO₂ was first used for the photocatalytic degradation of chlorinated biphenyls, semiconductor catalysts, with their highly efficient ability to degrade organic pollutants, have emerged as an emerging technology to address this issue, demonstrating considerable application potential^[18-20]. Due to the environmentally friendly semiconductor photocatalyst generating the powerful hydroxyl ($\cdot\text{OH}$) radicals for the oxidative decomposition of organic pollutants only depends on sunlight^[21], the photocatalytic oxidation process is favored by an increasing number of researchers^[22]. In the process of photocatalytic degradation, organic pollutants are decomposed into non-toxic and harmless inorganic substances, which promotes the sustainable utilization of resources^[23-25]. Take TiO₂ as an example, when sufficient light energy irradiates a TiO₂ catalyst, photogenerated e^-/h^+ pairs (with e^- being excited electrons and h^+ positively charged holes) are created between its valence and conduction bands. The h^+ has oxidation capacity and reacts with H₂O to produce $\cdot\text{OH}$. Meanwhile, the e^- has a reduced capacity and reacts with O₂ adsorbed on TiO₂ in a series of reactions to produce $\cdot\text{O}_2^-$ and $\cdot\text{OH}$.



where, $h\nu$ is a photon.

The photocatalytic oxidation process is extremely effective in degrading organic pollutants, however, several problems limit its application: (1) limited utilization of the solar spectrum^[26], so a large amount of visible light energy is wasted (the energy of the visible light used in the photocatalysis process, assuming a wavelength of 420–700 nm, is 2.84×10^{-19} – 4.73×10^{-19} J); (2) fast recombination rate of photogenerated carriers^[27,28]; (3) to improve catalytic efficiency, photocatalytic materials are usually nanoparticles, which makes them difficult to recycle. Meanwhile, some suspended matter in wastewater from livestock production will lead to deep chroma, which weakens light transmission and will inhibit photocatalytic activity. Therefore, it is necessary to improve the visible light capture ability and photocatalytic activity of photocatalytic materials to promote the photocatalytic oxidation process for wastewater treatment^[29]. In addition, semiconductor catalysts also have substantive effects in removing micro-pollutants such as antibiotics and drug residues from water, providing a new approach to solving the problem of micro-pollution in water bodies^[30,31].

In recent years, researchers have modified semiconductor catalysts through various methods to enhance their photocatalytic performance. For example, the band gap structure of semiconductor catalysts can be regulated through methods such as ion exchange, doping, recombination and heterojunction construction, thereby enhancing their absorption capacity for visible light^[32–34]. This review focuses on the development, application and catalytic efficiency of visible-light-driven TiO_2 -based, ZnO -based, and $g\text{-C}_3\text{N}_4$ -based (in which g is graphite) modified photocatalytic materials, as well as their application costs and prospects. We hope that this review paper can drive the development of highly efficient TiO_2 -based, ZnO -based, and $g\text{-C}_3\text{N}_4$ -based photocatalysts for treatment of wastewater from livestock production.

2 Mechanism of organic matter degradation via photocatalyst process

Most antibiotics in wastewater from livestock production are predominantly the following: tetracyclines, fluoroquinolones, sulfonamides, macrolides, chloramphenicols and β -lactams^[35–37]. Photocatalytic degradation can destroy the organic group of antibiotics and convert part of it to CO_2 and H_2O . Given the complexity of organic pollutants and the different photocatalytic materials used, there are different degradation pathways and intermediates even for the same pollutant in the photodegradation process. For example, in the photocatalytic degradation of oxytetracycline, which is a classic tetracycline antibiotic, there are 16 potential photocatalytic intermediates. Based on the charge-to-mass ratio ($m/z = 435, 431, 417, 416, 400, 379, 362, 347, 333, 319, 301, 275$ and 274), the formulas of intermediates could be obtained, which were presented in Fig. 1^[35]. In the $\text{TiO}_2/\text{Bi}_2\text{WO}_6/\text{rGO}$ (reduced graphene oxide) photocatalytic degradation of norfloxacin, the h^+ and $\cdot\text{OH}$ are the main reactive oxygen species^[36]. With the action of h^+ and $\cdot\text{OH}$, the norfloxacin is gradually broken down into small molecules of organic matter, mineralized inorganic matter, CO_2 , and H_2O (Fig. 2). When it comes to cephalixin (a classic cephalosporin antibiotic), which is widely used in human health and animal agriculture, ZnO nanowires were proven can degrade cephalixin effectively under simulated sunlight^[37]. The photocatalytic pathway of cephalixin by ZnO nanowires mainly included hydroxylation, demethylation, decarboxylation and dealkylation, and the specific degradation pathway is shown in Fig. 3.

Dyeing wastewater often has a deep chroma, similar to that of wastewater from livestock production, which can hinder light transmission. Taking the degradation of common azo-dyes as an example, methylene blue (MB) and rhodamine B (Rh B) can be completely degraded in $\text{MoS}_2/\text{MIL-88}(\text{Fe})$ system to form CO_2 , H_2O and other inorganic substances^[38]. The mineralization path of MB and Rh B dye was shown in Fig. 4, respectively. Under the action of the N-ethyl group mineralization, the colored $\text{C}=\text{N}$ bond breaks in the dyes structure, and N-demethylation occur. During N-demethylation, the $\text{C}-\text{C}$ and $\text{C}-\text{N}$ bonds of MB and Rh B are cleaved. Active oxidants are heavily involved in the degradation of MB and Rh B, which chemically convert major toxic pollutants into non-toxic secondary products such as water, carbon dioxide, and some mineral salts. Degradation of dyes is achieved by photocatalysts producing hydroxyl ($\cdot\text{OH}$) and

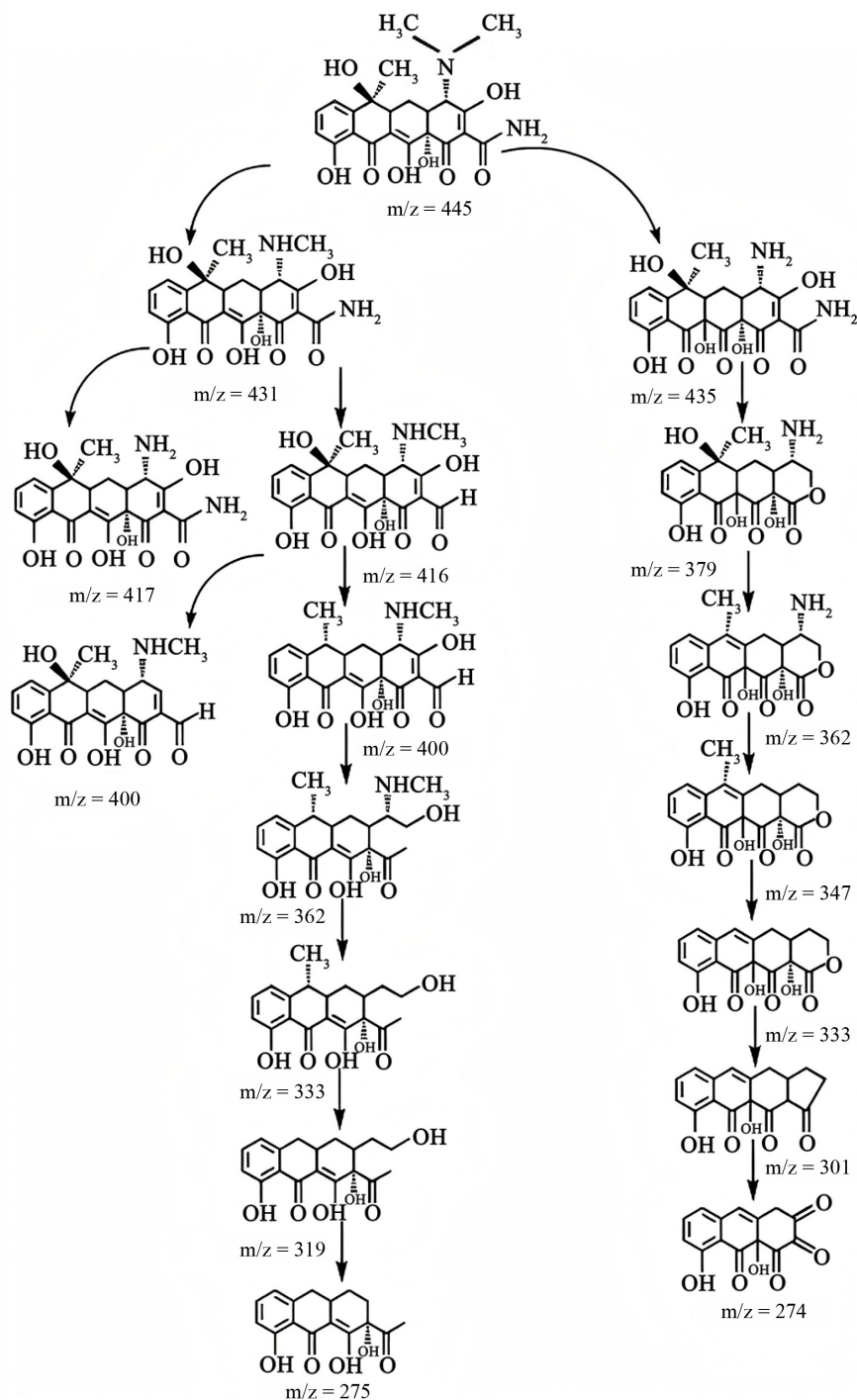


Fig. 1 Proposed pathways of photocatalytic tetracycline degradation. Reproduced from Huang et al.^[35], with permission from The Wiley-VCH Verlag GmbH & CO. KGaA.

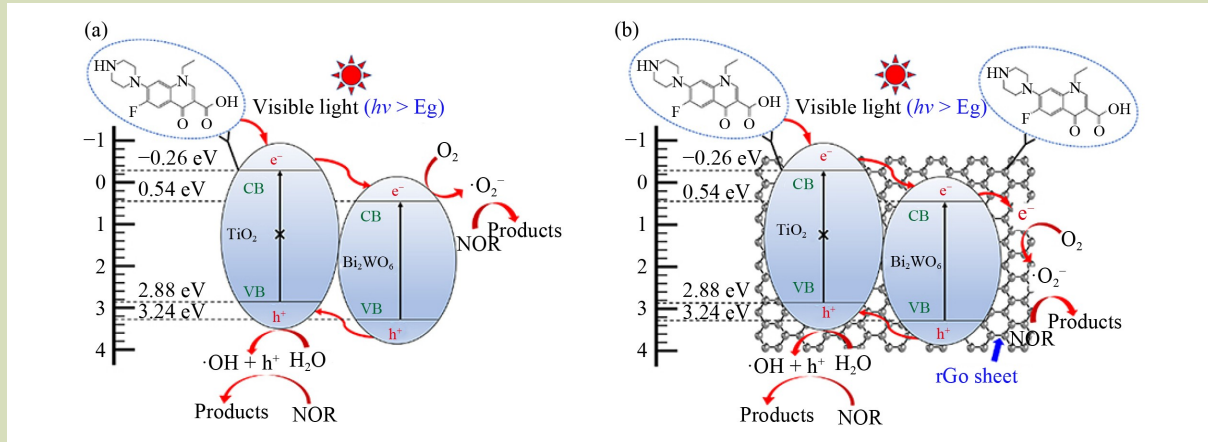


Fig. 2 The possible photocatalytic mechanism of degradation norfloxacin (NOR) by (a) TB and (b) TiO₂/Bi₂WO₆/rGO (reduced graphene oxide) under visible light irradiation. Reproduced from Ma et al.^[36], with permission from The Elsevier BV.

superoxide ($\cdot O_2^-$) under light. It can be seen that the modified photocatalyst has a well-catalytic effect even in wastewater with deep chroma.

3 Modification of photocatalysts

3.1 A brief introduction of TiO₂, ZnO and g-C₃N₄ photocatalyst

3.1.1 Titanium oxide

TiO₂ has excellent photocatalytic oxidation ability. For example, it has been reported that TiO₂ can photocatalytically degrade polychlorinated biphenyls, which are known for their hard-to-degrade nature, under UV light^[39]. This property has led to its widespread use in the field of photocatalysis, particularly in applications such as converting abundant solar energy into available hydrogen or hydrocarbon energy^[40,41], removing CO₂^[42] and photocatalytic degradation of organic pollutants^[43–46]. However, due to the forbidden band width of TiO₂ being 3.0–3.2 eV, the catalytic reaction can only take place under UV light ($\lambda < 400$ nm). As a result, its utilization rate for visible light is extremely low, typically less than 5%^[47]. Meanwhile, there is a certain attraction between photogenerated electrons and holes, and some of them recombine on the semiconductor surface, releasing heat instead of free radicals^[47]. This recombination process can reduce the quantum efficiency of TiO₂ photocatalysts to below 10%^[48]. In this case, the ability of TiO₂ photocatalyst to catalyze the

degradation of organic pollutants will be greatly reduced. Therefore, researchers have developed different modification methods to improve the photocatalytic capacity^[48].

3.1.2 Zinc oxide

ZnO, with the same band structure as TiO₂, is rich in source, low in price, diversified and adjustable in morphology and has suitable electrical conductivity, thermal conductivity and chemical stability. It is an environmentally-friendly wide-band-gap semiconductor photocatalyst^[49–51]. However, the ZnO band gap width of 3.37 eV means its utilization of sunlight is extremely low. For example, its visible light utilization rate is typically $< 3\%$ ^[52]. This limits its light energy utilization efficiency and catalytic activity to a certain extent. The photocatalytic capacity of ZnO remains suboptimal due to rapid electron-hole recombination, with recombination rates as high as 80%^[53], and limited visible light absorption^[54,55].

3.1.3 Graphitic carbon nitride

g-C₃N₄ is a typical polymer semiconductor with a band gap width of 2.7 eV. It can absorb the solar spectrum wavelength of less than 475 nm blue-violet light^[56]. The g-C₃N₄ has an appropriate semiconductor band edge position, which is different from TiO₂ and ZnO. It can effectively activate oxygen molecules and generate superoxide radicals for photocatalytic conversion of organic functional groups and degradation of organic pollutants^[57]. Meanwhile, the advantages of low raw material cost, easy synthesis, high stability, unique electronic

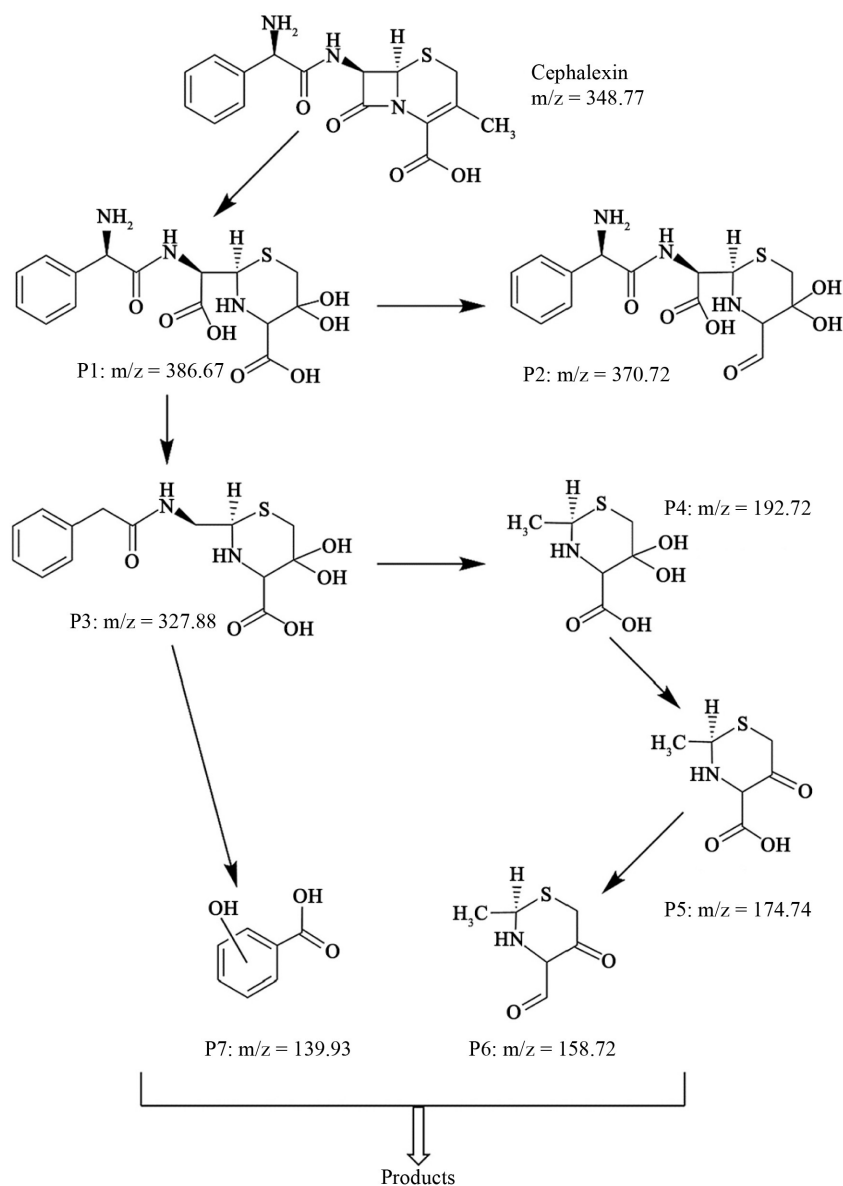


Fig. 3 Proposed photocatalytic degradation pathways of cephalosporin by ZnO nanowires under simulated sunlight irradiation. Reproduced from He et al.^[37] under Creative Commons.

structure and easy regulation make g-C₃N₄ a promising semiconductor photocatalyst^[58]. In addition, g-C₃N₄ has disadvantages such as a low utilization rate of visible light, typically around 10%–15%^[59] and a high composite probability of photogenerated electron holes, with recombination rates reaching up to 70%^[60]. To improve the performance of g-C₃N₄, a large number of modification studies have been carried out^[59,60].

3.2 Photocatalyst modification method

3.2.1 Morphology control

The structure of the photocatalyst itself is often decisive for catalytic efficiency. For example, TiO₂ mainly has three crystal structures anatase, rutile and plate titanium. Of these, the anatase lattice has the most defects and dislocations, which is more conducive to the generation of oxygen vacancy electron

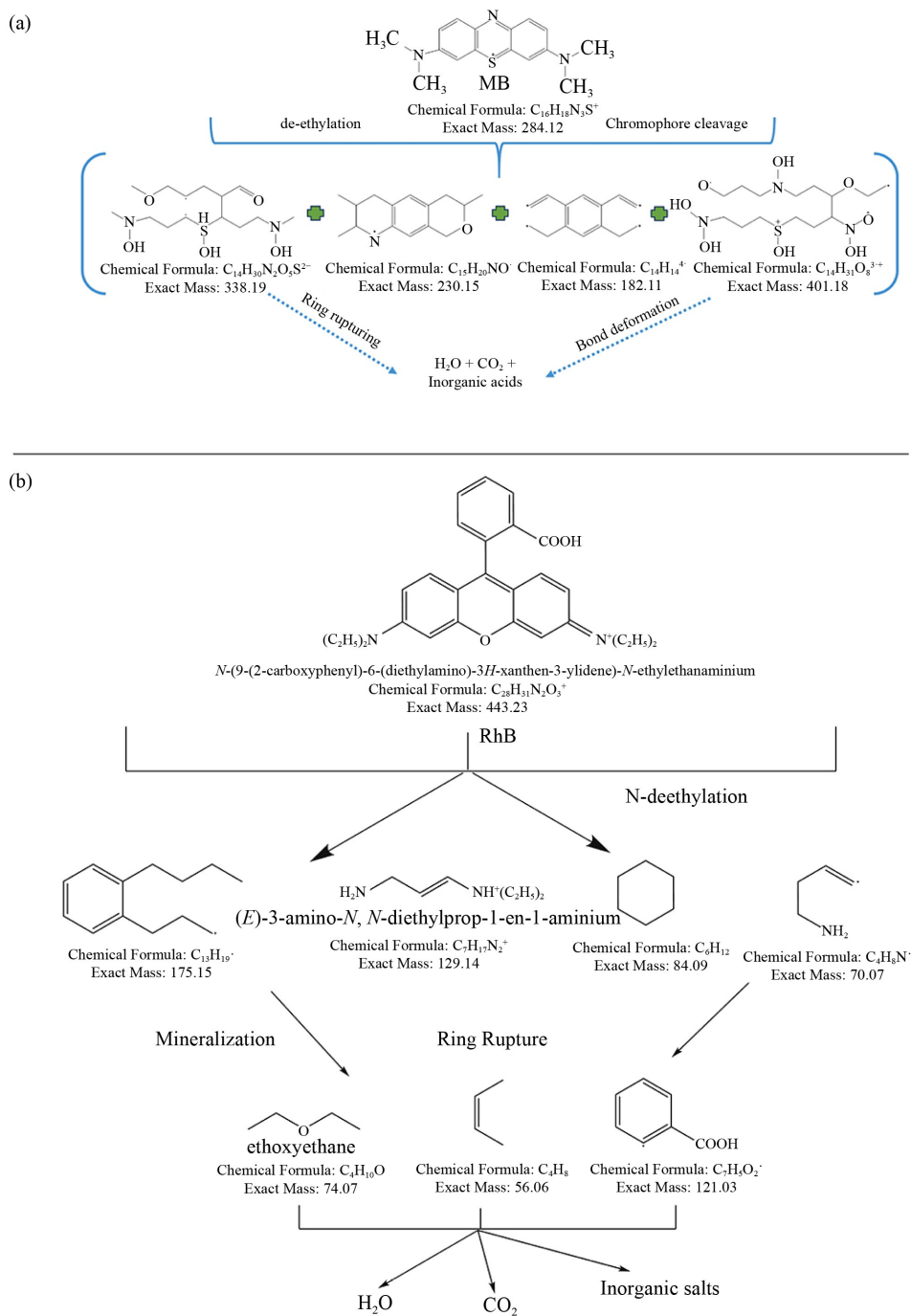


Fig. 4 (a) Proposed degradation pathway of methylene blue (MB) on $MoS_2@MIL-88(Fe)$ catalyst under visible light irradiation, (b) Proposed degradation pathway of rhodamine B (Rh B) on $MoS_2@MIL-88(Fe)$ catalyst under visible light irradiation. Reproduced from Govarthanan et al.^[38], with permission from The Elsevier BV.

capture and the inhibition of photoelectron-hole recombination. Therefore, anatase TiO₂ has a higher photocatalytic activity^[61]. The mixed crystal structure photocatalytic activity will be higher than that of single crystals when the anatase phase and rutile phase are calcined into TiO₂ mixed crystals in appropriate proportions^[62]. The main reason for this is that the crystal structure is equivalent to the composite of two kinds of semiconductors, which promotes the effective separation of photogenerated electrons and holes (Fig. 5).

The TiO₂ nanoparticles (anatase/rutile mixed crystals) could be used for the degradation of the hazardous dye MB under ultraviolet light illumination^[63]. pH1.0-TiO₂, which was synthesized under pH of 1.0, has a rod-like particle morphology and some irregularly shaped particles (Fig. 6). The research result shows that the mixed-phase TiO₂, which was

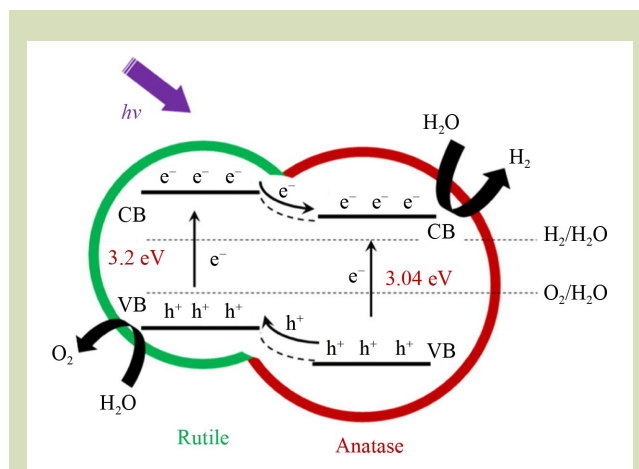


Fig. 5 Photocatalytic overall water splitting mechanism of TiO₂ nanobelts. Reproduced from Jacob et al.^[62], with permission from The Academic Press Inc.

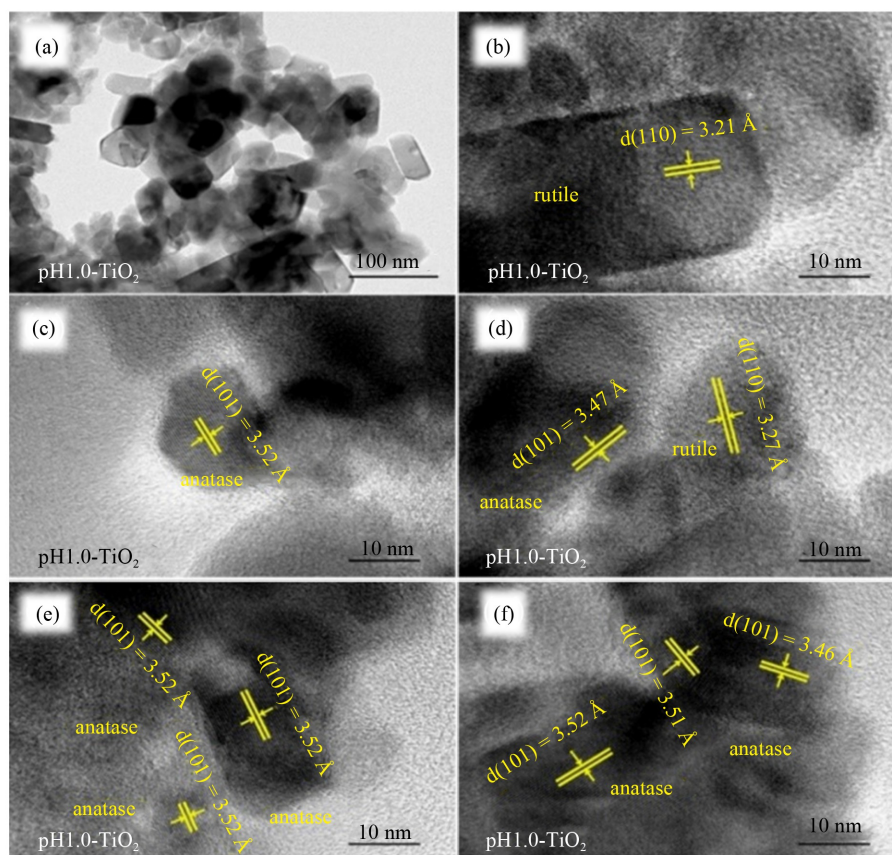


Fig. 6 Transmission electron microscopy (a) and high-resolution transmission electron microscopy (b-f) images of the TiO₂ sample obtained at pH 1.0 under hydrothermal conditions. Reproduced from He et al.^[63] under Creative Commons.

synthesized at pH of 1.0, has the highest photocatalytic activity of degradation MB is 85.8% under UV irradiation for 90 min. Compared with pH1.0-TiO₂, the degradation efficiency of the commercial P25 TiO₂ (Nippon Aerosil Ltd., Tokyo, Japan) to MB was only 79.4% under the same light condition.

3.2.2 Particle size control

With the decrease of the semiconductor nanoparticles size, band structure gradually transitions to the energy level structure of atoms or molecules. When the particle size is smaller than the space charge layer thickness, photoproduction carriers from internal migration to the surface more easily. It makes the light born in a semiconductor electron and hole not compound before it has arrived at the surface of the semiconductor, effectively inhibiting the photoproduction of the electronic-hole compound, and improving the quantum efficiency^[64,65]. As mentioned above, nano-ZnO can significantly improve the catalytic capacity of ordinary ZnO. This is because the smaller the particle size of photocatalyst nanoparticles, the larger the number of atoms on the surface, the larger the specific surface area, and the higher the light

absorption efficiency and adsorption efficiency of the catalyst surface, thus significantly improving the photocatalytic activity of nanomaterials^[66-68]. TiO₂ and g-C₃N₄ also have similar properties, which are similar to ZnO. The band gap shifted and carriers migrated to the surface rapidly with the decrease in particle size^[69]. When the particle size was as small as 2 nm, there would be an obvious quantum size effect. Using the electrochemical etching process to prepare ultrafine TiO₂ nanoparticles (with an average particle crystal size of 9.87 nm) exhibited high photocatalytic activity, which the photocatalytic degradation efficiency of tetracycline (TC) is 99.4%^[70].

The bulk g-C₃N₄ can be etched into nanosheets, which makes the photocatalytic activities significantly enhanced. Scanning electron microscope images confirm that the carbon nitride nanosheets combined with carbon quantum dots (CQDs) are a single-layer nanosheet with jagged edges and lateral dimensions ranging from submicron to several microns (Fig. 7)^[71]. In this particular construction, the substrate curvature makes the surface energy of materials minimizes, which enhances the photocatalytic activity of the catalysts.

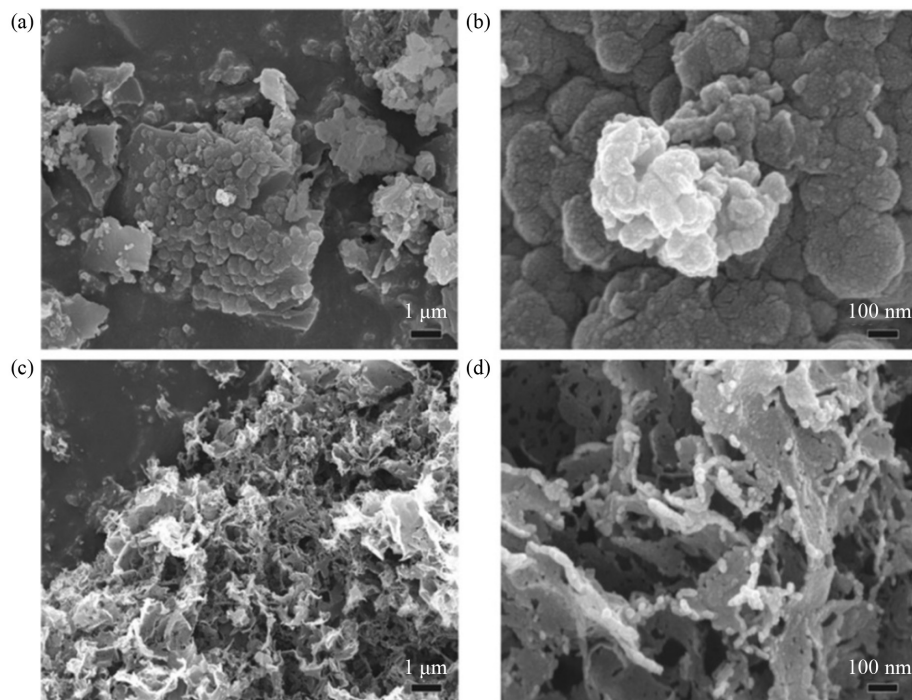


Fig. 7 Scanning electron microscopy images of (a, b) carbon nanobags and (c, d) carbon nitride nanosheets combined with carbon quantum dots. Reproduced from Wang et al.^[71], with permission from The Elsevier BV.

3.2.3 Increase specific surface area

The specific surface area of a material is defined as the total surface area of the material divided by its mass or volume. The degradation of organic pollutants by semiconductor photocatalytic oxidation occurs on the surface of the catalyst. With the increase of specific surface area, the formation of photocarriers and the adsorption and transport of reactants and degradation products on the surface are facilitated when other influencing factors, such as lattice defects on the catalyst surface, are the same^[72–74]. The optical effect of light sources on the surface of photocatalysts, such as multiple diffraction and diffuse reflection, increases the utilization rate of light and can effectively improve photocatalytic activity. The main ways to improve the specific surface area are the preparation of porous structure materials^[75–78], array structure^[79,80] and hierarchical structure^[81–83].

Anatase TiO₂ with a hollow hexagonal frame structure considerably increases its specific surface area^[75]. It is worth noting that the temperature and time of calcination are particularly critical in the process of material synthesis. As shown in Fig. 8(a), when the precursor was calcinated at 100 °C

for 2 h, the hexagonal smooth surfaces of TiO₂ crystals began to disappear. When the calcination temperature was raised to 300 °C, as shown in Fig. 8(b), the side faces of the hexagonal smooth surfaces disappeared and six edges gradually became apparent. However, the side faces of the hexagonal smooth surfaces kept their original morphology. It is easy to see that with the increase of calcination temperature, the TiO₂ crystals gradually grew into hexagonal box structures. When the calcination temperature is increased to 600 °C, it can be seen from the scanning electron microscopy image (Fig. 8(c)) that the material remained in hexagonal boxes. When the precursor was calcined (600 °C for 7 h), the prepared TiO₂ with a unique hexagonal framework structure, which has a higher specific surface area (51.9 m²·g⁻¹) than other calcination conditions. Using this material, with about threefold the surface area of its precursor when calcined at 100 °C for 2 h, increased the removal efficiency of Rh B solution to 98.6% within 40 min. The special array structure can also effectively modify the photocatalytic material. The hexagonal wurtzite nanorod arrays (polyimide/Ag)/ZnO-Ag can be prepared on polyimide, and polyimide/Ag nanofibers by combining electrospinning and hydrothermal reaction processes^[80]. The (polyimide/Ag)/ZnO-Ag had improved photocatalytic degradation of MB dye,

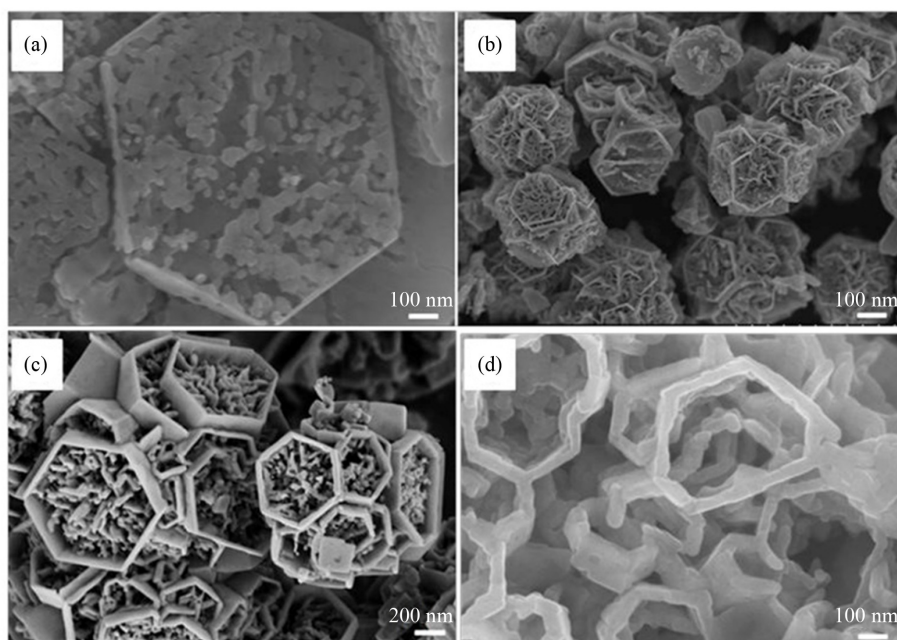


Fig. 8 Scanning electron microscopy images of TiO₂ synthesized with different calcined temperatures: (a) 100 °C for 2 h; (b) 300 °C for 2 h; (c) 600 °C for 2 h; (d) 600 °C for 7 h. Reproduced from Teng et al.^[75], under Creative Commons.

with a removal efficiency of 98% under illumination for 120 min.

The ability of g-C₃N₄ with the hierarchical porous structure to degrade organic pollutants is considerable. Using monodisperse SiO₂ as a template, g-C₃N₄ with a graded porous structure can be prepared by simple one-step calcination^[81]. The g-C₃N₄ prepared has a specific surface area and visible light absorption performance higher than bulk g-C₃N₄ which helps to facilitate the separation of photogenerated electron holes. The removal efficiency of MO of the catalyst with the best ratio of SiO₂ to dicyandiamide (1:1) reaches 60% within 100 min, which is threefold that of bulk g-C₃N₄. The existence of hierarchical microporous and mesoporous structures can provide additional reaction sites for photocatalytic reactions, which is conducive to enhancing catalytic performance.

3.3 Main modification methods for photocatalysts

3.3.1 Elements-doped

(1) Metal ion-doped

Modification refers to the adjustment of the structure, composition or surface properties of materials through physical or chemical methods to improve their performance. Element doping refers to the introduction of a small number of heterogeneous atoms into materials to alter their electronic structure and physicochemical properties, thereby enhancing the performance of the material.

Metal ions used for semiconductor doping mainly include transition metal ions (Fe³⁺, Co²⁺ and Cu²⁺), rare earth metal ions (La³⁺, Ce³⁺ and Pr³⁺), and inorganic functional group ions

[Fe (CN)₆⁴⁻ and MoS₄²⁻], as shown in Table 1. Of these, transition metal ions and rare earth metal ions are the most common^[82–87].

TiO₂ nanosheets, with Cu²⁺ doping by solvothermal synthesis, have a high specific surface area and excellent visible light response^[82]. By introducing Fe³⁺, the light absorption range can be further extended from the ultraviolet band to the visible range^[83] (Fig. 9). The removal efficiency of Fe³⁺ doped TiO₂ nanotube arrays to methyl orange (MO) was about 1.5 times higher than that of pure TiO₂ nanotube arrays within 120 min. Loading metal ions elements on the surface of nano-ZnO can also provide useful improvement^[84–86]. Khan et al.^[85] prepared Fe³⁺/ZnO photocatalytic materials by sol-gel synthesis method. The photocatalytic activity was tested to degrade 4-chlorophenol under visible light, Fe-ZnO of optimum loading proportion had the highest activity, degrading 73% 4-chlorophenol. Another study^[86] also found that the Co²⁺ could effectively tune the electronic and optical properties of ZnO in photocatalytic applications. When doped with 3 mol% Co²⁺, Co²⁺/ZnO exhibited the best catalytic effect and the degradation efficiency reached 99.7% of direct blue 71 in 150 min. Compared with TiO₂ and ZnO, g-C₃N₄ has a higher activation efficiency for molecular oxygen, which is more conducive to the catalytic degradation of organic pollutants. Under visible light irradiation, the porous Fe³⁺/g-C₃N₄ can almost complete the degradation of sulfadiazine with a removal efficiency as high as 99.8% within 90 min^[87].

(2) Non-metallic elements-doped

In general, doping of non-metallic elements such as C, N, B, S and F positively influences the photography of semiconductors in the visible region^[88–95], as shown in Table 2. The

Table 1 Summary of the results for metal ion-doped photocatalyst systems

Photocatalyst and dosage	Optimum doping ratio	Organic pollutant and concentrations	Photodegradation efficiency	Ref.	
TiO ₂	Cu/TiO ₂ (30 mg)	4% Cu doping amount	[TC] = 30 mg·L ⁻¹	100 min, over 90%	[82]
	Fe ³⁺ /TiO ₂ (thin film)	0.2 mol·L ⁻¹ Fe ³⁺ doping	[MO] = 2 mg·L ⁻¹	120 min, over 70%	[83]
	Ce/ZnO (150 mg)	–	[RO29] = 10 mg·L ⁻¹	50 min, 97.84%	[84]
ZnO	Fe ³⁺ /ZnO (800 mg)	0.8% Fe ³⁺ doping amount	[4-CP] = 0.6 mg·L ⁻¹	60 min, 73%	[85]
	Co ²⁺ /ZnO (100 mg)	Co ²⁺ ion of 3 mol%	[DB71] = 100 mg·L ⁻¹	150 min, 99.7%	[86]
g-C ₃ N ₄	Fe ³⁺ /g-C ₃ N ₄ (0.1 mg·L ⁻¹)	3.2% Fe ³⁺ doping amount	[SDZ] = 2 mg·L ⁻¹	90 min, 99.8%	[87]

Note: “–”, not given in the reference.

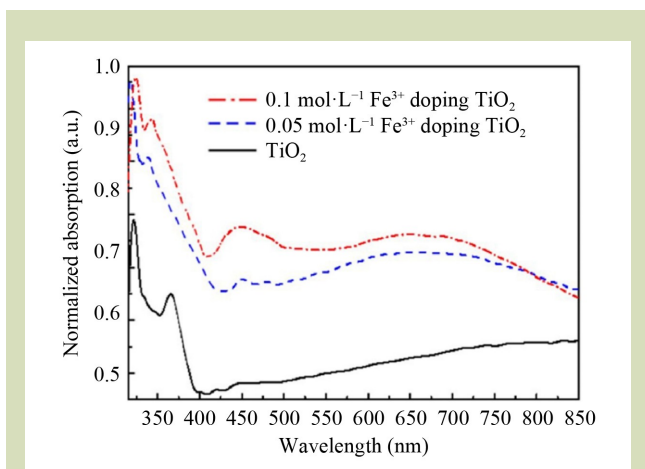


Fig. 9 UV-Vis absorption spectra of pure and Fe³⁺ doped TiO₂ nanotube arrays. Reproduced from Zhang et al.^[83] under Creative Commons.

mechanism of improving photocatalyst modification by doping non-metallic elements has not yet been determined. Taking N-doped TiO₂ as an example, the reason can be simply explained as N-doped causes oxygen vacancy and enhances photocatalytic performance. The N-TiO₂ catalyst, obtained after baking in an ammonia atmosphere for 1 h with abundant oxygen vacancies, provides photocatalytic activity 7.59 and 2.26 times higher than pure TiO₂ and the commercial P25 TiO₂ (Evonik Industries)^[89], respectively. The N-doped ZnO photocatalyst also has a good photocatalytic effect under visible light. The N-doped ZnO photocatalyst has been synthesized by sol-gel method and photocatalysis experiments show that N-doped ZnO has a significant enhancement of degradation for

MB under irradiation of both UV and visible light region^[91]. Similarly, N self-doping enhances the utilization of visible light by g-C₃N₄ nanosheets, promotes photogenerated electron-hole separation, and prolonged the lifetime of photogenerated charge carriers^[94,95]. As N atoms occupy oxygen vacancies, the band gap width of the photocatalytic material was reduced and the motion of photogenerated electrons was accelerated, which in turn facilitates the photocatalytic reaction.

3.3.2 Noble metal deposition

The photocatalytic reaction takes place on the semiconductor surface, so its surface structure has a considerable influence on photocatalytic performance. Noble metals have higher corrosion resistance and oxidation resistance, which enables photocatalysts remain stable during the catalytic process. In addition, the deposition of noble metal on the surface of photocatalytic material can change the surface properties of the catalyst and the electron distribution of the system, to improve the photocatalytic performance^[96-101], as shown in Table 3.

Ag has excellent electrical conductivity, so it is often used as a noble metal element in modified photocatalysts. Podasca and Damaceanu^[98] synthesize hybrid polymer films loaded with ZnO-Ag particles and explore their photocatalytic removal efficiency of MO. Its degradation efficiency for MO peaked at 95% when a hybrid film was loaded with 5% ZnO-Ag particles. Due to the synergistic effect between noble metals, the deposition of two noble metals tends to provide better modification of photocatalytic materials. Lee et al.^[100] prepared ZnO nanocomposites modified with bimetallic Au and Pd nanoparticles by poly(lactic acid) technique and photodeposition

Table 2 Summary of the results for non-metallic elements-doped photocatalyst systems

Photocatalyst and dosage		Optimum doping ratio	Organic pollutant and concentrations	Photodegradation efficiency	Ref.
TiO ₂	S/TiO ₂ (30 mg)	S ⁴⁺ /S ⁶⁺ ratio of 1:5.5	[CIP] = 10 mg·L ⁻¹	140 min, 91.5%	[88]
	N/TiO ₂ (50 mg)	1 h in an NH ₃ atmosphere	[TC] = 20 mg·L ⁻¹	10 min, over 80%	[89]
	C/TiO ₂ (100 mg)	2 atoms% of carbon	[MB] = 20 mg·L ⁻¹	120 min, over 90%	[90]
ZnO	N/ZnO (10 mg)	2 h under N ₂ atmosphere	[MB] = 10 mg·L ⁻¹	180 min, 80%	[91]
	S/ZnO (100 mg)	0.5 wt% S-doped	[Rh B] = 5 mg·L ⁻¹	90 min, 100%	[92]
g-C ₃ N ₄	Cl/g-C ₃ N ₄ (50 mg)	NH ₄ Cl to melamine is 1:1	[TC] = 10 mg·L ⁻¹	120 min, 92%	[93]
	N/g-C ₃ N ₄ (50 mg)	-	[TC] = 10 mg·L ⁻¹	60 min, 81.72%	[94]
	N/g-C ₃ N ₄ (100 mg)	N to CN is 2 mg:1 g	[Phenol] = 10 mg·L ⁻¹	180 min, 70.1%	[95]

Note: “-”, not given in the reference.

Table 3 Summary of the results for noble metal deposition photocatalyst systems

Photocatalyst and dosage		Optimum doping ratio	Organic pollutant and concentrations	Photodegradation efficiency	Ref.
TiO ₂	Pd/TiO ₂ (8 mg)	0.5 wt% of Pd	[PCB 3] = 0.2 mol·L ⁻¹	30 min, 100%	[96]
	Ag/TiO ₂ (25 mg)	5% of 30 nm Ag	[TC] = 10 mg·L ⁻¹	90 min, 90%	[97]
ZnO	Ag/ZnO (1 g)	5 wt% of Ag	[MO] = 1 × 10 ⁻³ mol·L ⁻¹	250 min, 95.17%	[98]
	Nd-V-ZnO (180 mg)	Nd (4 mol%) and (1 mol%) V	[MO] = 10 mg·L ⁻¹	150 min, 99.3%	[99]
	Au/Pd/ZnO (0.5 mg)	Au 15 wt% and Pd 5 wt%	[MB] = 5.0 × 10 ⁻⁴ mol·L ⁻¹	180 min, 97%	[100]
g-C ₃ N ₄	Au/g-C ₃ N ₄	1 wt% of Au	[Paracetamol] = 0.3 mg·L ⁻¹	25 min, 100%	[101]

method, and the ZnO/Au/Pd nanocomposite removal efficiency of MB was about 5.4 times than pure ZnO.

Although noble metal deposition can considerably enhance photocatalytic performance, inevitably increasing the raw material cost of preparing photocatalytic materials, it greatly restricts the development and application of this kind of material. The search for substitutes is extremely necessary. There are several non-noble metal alternatives that have shown promising performance for specific pollutants. Copper has been frequently investigated as an alternative to rare metal materials, especially in catalysis. For example, Cu-based nanomaterials have demonstrated excellent catalytic efficiency in the oxidation and decomposition of organic pollutants, providing cost-effectiveness far superior to that of precious metal catalysts^[102]. Also, a Cu/TiO₂ composite has been reported to have excellent activity in the degradation of organic pollutants^[103]. Also, Mo, having an elemental abundance that lies between that of the non-noble and the noble metals and a price comparable to that of non-noble metals, has been examined as an alternative to noble metals^[104,105]. For example, MoS₂ nanoflowers have provided suitable photodegradation performance for various pollutants^[106]. In terms of photocatalysts, two-dimensional materials, carbon-based quantum dots and Mxenes are emerging as promising alternatives.

3.3.3 Semiconductor composite photocatalysts

Semiconductor compositing is a common modification method to improve the photocatalytic capacity. By combining two or more materials under the premise of an appropriate energy band to modify the photocatalyst, the composite material not only effectively adjusts the performance of a single material, but also generates many new photochemical and

physical properties. Binary compound semiconductors, for example, with two types of semiconductor compounds of different band structure, have potential differences that promote the separation of electron-hole pair^[107-123] (Table 4).

Combining the two kinds of semiconductors to form semiconductor composite photocatalysts can also improve visible light utilization. The MoS₂@TiO₂ nanosheet heterojunction composite material was used for photocatalytic degradation of MB, with highest removal efficiency of 86% within 180 min, which is much greater than that of pure TiO₂^[107]. When the two semiconductors are compounded, their energy band structure is also changed, which improves the absorption and utilization of visible light and decreases the recombination rate of the photogenerated electron-hole. One study^[109] also found that SnIn₄S₈ nanosheet/TiO₂ hollow sphere heterojunction has high photocatalytic efficiency. When the addition of SnIn₄S₈ increases, the light absorption range of the SnIn₄S₈/TiO₂ heterostructure broadens toward the visible band, which means that the addition of SnIn₄S₈ helps to enhance the absorption of visible light by TiO₂. The NaBiS₂ is a narrow-band-gap semiconductor. When it is combined with a wide-band-gap semiconductor, like ZnO, it can enhance the oxidation ability and photocatalytic activity^[112]. NaBiS₂/ZnO nanocomposites provide greater photocatalytic ability than pure NaBiS₂ and ZnO. The removal efficiency of the optimal ratio NaBiS₂/ZnO nanocomposite for Rh B is 99% under visible light irradiation within 120 min. The transfer of photogenerated electrons through the interface between NaBiS₂ and ZnO slows down the recombination rate of electron-hole pairs, which improves the efficiency of photocatalysis. The g-C₃N₄-based composite photocatalysts have a strong photocatalytic capacity for organic pollutants in wastewater under visible light. One study^[121] has reported that CoP as a co-catalyst modified g-C₃N₄ (HCCN) to form a stable

Table 4 Summary of the results for semiconductor composite photocatalyst systems

Photocatalyst and dosage		Optimum doping ratio	Organic pollutant and concentrations	Photodegradation efficiency	Ref.
TiO ₂	MoS ₂ /TiO ₂ (A thin film)	–	[MB] = 5 mg·L ⁻¹	180 min, 86%	[107]
	Black anatase-TiO ₂ (20 mg)	–	[TC] = 10 mg·L ⁻¹	270 min, 66.2%	[108]
	SnIn ₄ S ₈ /TiO ₂ (20 mg)	5 wt% of SnIn ₄ S ₈	[MO] = 15 mg·L ⁻¹	60 min, 90.1%	[109]
	Bi ₂ WO ₆ @TiO ₂ (10 mg)	–	[MB] = 10 mg·L ⁻¹	70 min, 96%	[110]
	CoFe ₃ O ₄ @TiO ₂ (5 mg)	–	[MB] = 100 mg·L ⁻¹	60 min, 91%	[111]
ZnO	NaBiS ₂ /ZnO (10 mg)	NaBiS ₂ /ZnO is 10:1	[Rh B] = 50 mL	120 min, 99%	[112]
	Bi ₂₄ O ₃₁ Br ₁₀ /ZnO (50 mg)	5 wt% of Bi ₂₄ O ₃₁ Br ₁	[Rh B] = 10 mg·L ⁻¹	40 min, 92%	[113]
	Ag ₈ S/ZnO (50 mg)	–	[Rh B] = 1 × 10 ⁻² mol·L ⁻¹	120 min, over 90%	[114]
	AgIO ₄ /ZnO (100 mg)	15 wt% of AgIO ₄	[IC] = 5 × 10 ⁻⁵ mol·L ⁻¹	180 min, 88%	[115]
	g-C ₃ N ₄ /ZnO (25 mg)	–	[Crystal violet] = 20 mg·L ⁻¹	180 min, 97%	[116]
g-C ₃ N ₄	CuInS ₂ /g-C ₃ N ₄ (50 mg)	50 wt% of CuInS ₂	[TC] = 20 mg·L ⁻¹	60 min, 83.7%	[117]
	Bi ₄ O ₅ I ₂ /g-C ₃ N ₄ (100 mg)	–	[MO] = 20 mg·L ⁻¹	40 min, 99.96%	[118]
	Bi ₇ O ₉ I ₃ /g-C ₃ N ₄ (50 mg)	mass ratio of Bi (NO ₃) ₃ ·5H ₂ O/g-C ₃ N ₄ is 12.125	[DOX] = 20 mg·L ⁻¹	120 min, 80%	[119]
	WO ₃ /g-C ₃ N ₄ (50 mg)	–	[TC] = 20 mg·L ⁻¹	120 min, 90.54%	[120]
	CoP/g-C ₃ N ₄ (40 mg)	5 wt% of CoP	[TC] = 10 mg·L ⁻¹	120 min, 96.7%	[121]
SnO ₂ /g-C ₃ N ₄ (50 mg)	3 wt% of SnO ₂	[TC] = 30 mg·L ⁻¹	120 min, 95.90%	[122]	
CoFe ₂ O ₄ /g-C ₃ N ₄ (200 mg)	3 wt% of CoFe ₂ O ₄	[4-CP] = 1 × 10 ⁻³ mol·L ⁻¹	240 min, about 50%	[123]	

Note: “–”, not given in the reference.

and highly efficient CoP/HCCN composite via a simple solvothermal method. The semiconductor composite photocatalyst provided highly efficient catalytic degradation of TC. With CoP/HCCN of 5 wt% of CoP, TC was degraded by 96.7% within 120 min, which was 10.2 times higher than the degradation efficiency of TC by HCCN.

Although semiconductor composite photocatalysts provide suitable catalytic performance and controllable cost, the narrow-band-gap semiconductors which are matched with TiO₂, ZnO and g-C₃N₄ are often toxic, difficult to prepare, or unstable and prone to photo corrosion. Therefore, further development of low-cost, non-toxic, and harmless narrow-band-gap semiconductors is key to promoting the large-scale application of semiconductor composite photocatalysts.

3.3.4 Dye-sensitized and quantum dot-sensitized

Dye-sensitized and quantum dot-sensitized are new methods to improve visible light catalytic degradation of organic pollutants with TiO₂, ZnO and g-C₃N₄-based

photocatalysts^[124–128]. Table 5 shows several common dye-sensitized and quantum dot-sensitized methods reported in recent years.

Dyes adsorbed on semiconductor surfaces can absorb all visible light and even near-infrared light. As visible light is absorbed by dye, the electrons on the dye will jump from the ground state to the excited state. When the potential of the free electrons generated by the excited dye is higher than the potential of the photocatalytic materials conduction band, the electrons on the dye will be transferred to the semiconductor. Using organic dye D35 sensitized TiO₂ nano-crystalline film to form a high-efficient visible-light photocatalyst, which has over 85% degradation efficiency of bisphenol A (BPA) within 180 min^[124]. The study also found that using chlorophyll sensitized TiO₂ nanoparticles photocatalytic can efficiently degrade MB under visible light, which optimum removal efficiency was 85% within 2 h^[125]. The key to dye-sensitized lies in the rapid injection of excited electrons into the semiconductor photocatalyst, and avoiding the recombination of excited electrons and dye positive ions free radicals.

Table 5 Summary of the results for dye-sensitized and quantum dot-sensitized photocatalyst systems

Photocatalyst and dosage		Optimum doping ratio	Organic pollutant and concentrations	Photodegradation efficiency	Ref.
TiO ₂	D35 Dye-sensitized/TiO ₂ (8 mg)	0.1 mmol·L ⁻¹ D35-dye	[BPA] = 5 mg·L ⁻¹	180 min, over 85%	[124]
	chl Dye-sensitized/TiO ₂ (250 mg)	0.1 wt% of chl	[MB] = 20 mg·L ⁻¹	120 min, 85%	[125]
	CQDs/TiO ₂ (4 mg)	Weight ratio of 1:10 of CQD and TiO ₂	[MO] = 20 mg·L ⁻¹	180 min, over 80%	[126]
g-C ₃ N ₄	CQDs/g-C ₃ N ₄ (10 mg)	with CQDs stock solution 50 μL	[DCF] = 10 mg·L ⁻¹	60 min, 100%	[127]
	BPQDs/g-C ₃ N ₄ (30 mg)	-	[OTC and Rh B] = 10 mg·L ⁻¹ [TC] = 20 mg·L ⁻¹	60 min, 81.05% (OTC) 20 min, 99.43% (Rh B) 60 min, 77.11% (TC)	[128]

Note: “-”, not given in the reference.

Quantum dots are nanoscale semiconductors, and the principle of quantum dot-modified photocatalyst is to use light energy to excite the electrons on the surface of carbon quantum dots to form electron-hole pairs, thus promoting the photocatalytic reaction process^[126–128]. CQDs are zero-dimensional, fluorescent carbon-based nanomaterials, typically smaller than 10 nm in size with a substantial percentage of oxygen and hydrogen atoms on their surface, which gives them low toxicity and excellent biocompatibility. CQD sensitizing is a promising way to enhance the efficiency of degraded organic pollutants of photocatalysts under visible light. CQD/TiO₂ provides high-efficiency MO photodegradation in direct sunlight exposures, which the optimized doping ratio in the weight ratio of CQD and TiO₂ (threefold the activity of pure TiO₂)^[126]. The CQD modified g-C₃N₄ with optimal CQDs loading exhibits extremely high photocatalytic efficiency, which has 15 times the removal efficiency to diclofenac higher than that of pure g-

C₃N₄^[127]. In addition to CQDs, BPQDs can also have the same effect. The tubular g-C₃N₄ with BPQDs has unusual photocatalytic efficiency in the degradation of oxytetracycline hydrochloride (0.0276 min⁻¹), which is 4.78 times more than CN and 2.36 times more than tubular g-C₃N₄^[128].

3.3.5 Compound modification

Various modification methods can effectively enhance the ability of photocatalysts to degrade organic pollutants in visible light, but there are still some difficulties to overcome. Therefore, more and more methods are needed to modify photocatalytic materials^[13,108,129–158] (Table 6). For example, the research results show that nanomaterials can significantly improve the catalytic capacity compared with ordinary materials. Nanomaterials as catalysts have large specific surface area, contact area and useful active particle effect, which is the

Table 6 Summary of the results for compound modification photocatalyst systems

Photocatalyst and dosage		Organic pollutant and concentrations	Photodegradation efficiency	Ref.
TiO ₂	NP-Fe ₃ O ₄ /GO-TiO ₂ (80 mg)	[Rh B] = 20 mg·L ⁻¹	180 min, over 85%	[129]
	N-TiO ₂ @SiO ₂ @Fe ₃ O ₄ (50 mg)	[PPCPs] = 2 mg·L ⁻¹	5 h, 94% (IBU) 5 h, 93% (BZP-3) 9 h, 71% (CBZ) 9 h, 71% (CBZ)	[130]
	Fe ₃ O ₄ @SiO ₂ @TiO ₂ -Co/rGO (0.3 g)	[MB] = 10 mg·L ⁻¹	160 min, 98.9%	[131]
	S-N/CoFe ₂ O ₄ @rGO@TiO ₂ (80 mg)	[Organic dyes] = 5 mg·L ⁻¹	120 min, 100% (MO) 90% (MB); 65% (Rh B)	[132]
	S-Bi-F-TiO ₂ /SiO ₂ (500 mg)	[Acrylonitrile] = 10 mg·L ⁻¹	6 min, 81.9%	[133]
	CeO _x @C-TiO ₂ (50 mg)	[TC] = 40 mg·L ⁻¹	180 min, 83.5%	[134]
	Black anatase-TiO ₂ (20 mg)	[TC] = 10 mg·L ⁻¹	270 min, 66.2%	[108]

(Continued)

Photocatalyst and dosage		Organic pollutant and concentrations	Photodegradation efficiency	Ref.
ZnO	ZnO/N-rGO (100 mg)	[2,4-DCP] = 10 mg·L ⁻¹	60 min, over 50%	[135]
	ZnO/Fe ₃ O ₄ /g-C ₃ N ₄ (10 mg)	[Azo dyes] = 30 mg·L ⁻¹	150 min, 97.9% (MO); 98.1% (AYR); 83.4% (OG)	[136]
	ZnO/GO/Ag ₃ PO ₄ (50 mg)	[TC] = 30 mg·L ⁻¹	75 min, 96.3%	[137]
	Ag ₃ PO ₄ /g-C ₃ N ₄ /ZnO (30 mg)	[TC] = 30 mg·L ⁻¹	180 min, 90.0% (Sunlight) 88.5% (Vis-Light)	[138]
	Fe ₃ O ₄ /ZnO/Si ₃ N ₄ (20 mg)	[MO and SY] = 50 mg·L ⁻¹	90 min, 96% (MO) 90 min, 90% (SY)	[13]
	La-ZnO/SiO ₂ (15 mg)	[MG] = 15 mg·L ⁻¹	140 min, 96.1% (MG)	[139]
	ZnO/Cds/GO (50 mg)	[MO] = 20 mg·L ⁻¹	120 min, 100%	[140]
	AC@Au/ZnO (6 mg)	[Gemifloxacin] = 20 mg·L ⁻¹	35 min, 98.0%	[141]
	MnFe ₂ O ₄ /ZnO/CQDs (62.5 mg)	[Gentamicin] = 50 mg·L ⁻¹	120 min, 69.1%	[142]
	Ce/ZnO/Al ₂ O ₃ (23 g)	[AR88 azo dye] = 20 mg·L ⁻¹	240min, over 95%	[143]
	Fe-ZnO/WO ₃ (100 mg)	[LVF] = 10 mg·L ⁻¹	180 min, 96%	[144]
	ZnO/Zn ₂ TiO ₄ /GO (0.88 g·L ⁻¹)	[CEF] = 10 mg·L ⁻¹	115 min, 71.4%	[145]
	g-C ₃ N ₄	S-CQDs/g-C ₃ N ₄ (1 g·L ⁻¹)	[TC] = 20 mg·L ⁻¹	40 min, 82.7%
g-C ₃ N ₄ /PDI@NH ₂ -MIL-53(Fe) (10 mg)		[Organic pollutants] = 50 mg·L ⁻¹	60 min, 90% (TC) 150 min, 78% (CBZ) 10 min, 100% (BPA) 30 min, 100% (PNP)	[147]
g-C ₃ N ₄ /PDI@NH ₂ -MIL-53(Fe) (10 mg)		[Organic pollutants] = 50 mg·L ⁻¹	60 min, 90% (TC) 150 min, 78% (CBZ) 10 min, 100% (BPA) 30 min, 100% (PNP)	[147]
Ag/g-C ₃ N ₄ /Bi ₃ TaO ₇ (25 mg)		[SMZ] = 20 mg·L ⁻¹ [MO] = 5 mg·L ⁻¹	25 min, 98% (SMZ) 40 min, 89% (MO)	[148]
Ag ₃ PO ₄ /Co ₃ (PO ₄) ₂ /g-C ₃ N ₄ (50 mg)		[TC] = 10 mg·L ⁻¹	120 min, 88%	[149]
Bi ₂ MoO ₆ /g-C ₃ N ₄ /Au (5 mg)		[Rh B] = 5 mg·L ⁻¹	40 min, 97.6%	[150]
Cu ₃ P-ZnSnO ₃ /g-C ₃ N ₄ (50 mg)		[TC] = 10 mg·L ⁻¹	60 min, 98.45%	[151]
Ag-AgCl/WO ₃ /g-C ₃ N ₄ (50 mg)		[TMP] = 4 mg·L ⁻¹	30 min, 99.9%	[152]
CDs/g-C ₃ N ₄ /SnO ₂ (10 mg)		[IDM] = 10 mg·L ⁻¹	80 min, 90.8%	[153]
Ag-g-C ₃ N ₄ /SnS ₂ (20 mg)		[TC] = 15 mg·L ⁻¹	150 min, 94.9%	[154]
N/K ₄ Nb ₆ O ₁₇ /g-C ₃ N ₄ (100 mg)		[Rh B] = 1 mg·L ⁻¹	60 min, 100%	[155]
FeOOH NPs/Nvac-CNNS (g-C ₃ N ₄) (40 mg)		[OTC] = 10 mg·L ⁻¹	90 min, 92.8%	[156]
Ag ₃ PO ₄ /g-C ₃ N ₄ @MoS ₂ (30 mg)		[AMA] = 20 mg·L ⁻¹	90 min, 100%	[157]
TiO ₂ @g-C ₃ N ₄ @BC (200 mg)	[CIP] = 20 mg·L ⁻¹	60 min, 89.2%	[158]	

most outstanding characteristic. However, with the decrease in particle size, nanoparticle catalysts are not easy to disperse and difficult to recover, so it is a good solution to this problem to load the catalytic material with porous materials mainly composed of magnetic materials. Qi et al.^[129] prepared a novel magnetic GO-TiO₂ composite (Fe₃O₄/GO-TMC), which has a

removal efficiency for Rh B over 85% under visible light irradiation within 180 min. Meanwhile, for the as-synthesized Fe₃O₄/GO-TMC no obvious decline in degradation efficiency was observed after at least five cycles and it was easier to recycle due to magnetic material doping. In addition, there have several other magnetically supported composite photocatalysts

have been used for the degradation of pharmaceuticals and personal care products^[130], dyes^[131,132,136] and antibiotics^[142] with useful effects.

In addition to being easy to recycle, ternary heterojunctions provide better photocatalytic performance than standard catalysts. GO is a new type of carbon material with a two-dimensional honeycomb lattice structure, which has excellent mechanical, thermal and electrical properties. GOs can be combined with others photocatalytic materials to achieve synergistic treatment of wastewater pollutants^[135,137,140,145]. The research results revealed that the ZnO/GO/Ag₃PO₄ composite provides higher photocatalytic activity for TC degradation under visible light than pure Ag₃PO₄, and ZnO/GO/Ag₃PO₄, with 96.3% degradation efficiency of TC within 75 min. Compared to Ag₃PO₄, which lost significant photocatalytic efficiency after three cycles of use, ZnO/GO/Ag₃PO₄ maintained relatively high photocatalytic performance after three cycles, indicating the stability of the photocatalyst. Through GO, the photogenerated electrons can rapidly transfer from Ag₃PO₄ to ZnO, so efficiently avoid the reduction of Ag⁺^[137].

Biochar is a solid substance rich in carbon produced by the pyrolysis of biomass under the condition of oxygen isolation. It has a large specific surface area and diverse functional groups, offering excellent adsorption, electrical conductivity and chemical stability, making it widely used as a carrier. In addition, the excellent electrical conductivity of biochar contributes to the transfer of photogenerated electrons, which can effectively reduce the compounding rate of photogenerated electrons and holes on the photocatalysts surface, and thus improve the photocatalytic efficiency. Faisal et al.^[141] produce the ZnO doped by activated carbon (AC) and Au nanoparticles by the hydrothermal and ultrasonication methods. The high-efficiency AC@Au/ZnO framework provided impressive results with 98.0% destruction of the gemifloxacin mesylate drug, being 2.21 times higher than pure ZnO and 1.63 times than Au/ZnO. Wang et al.^[158] loaded TiO₂/g-C₃N₄ with bamboo biochar and used it to degrade the ciprofloxacin under visible light giving 89.2% with 60 min. This material also had suitable stability and high photocatalytic activity even after five cycles.

Loading hard to recover and easily aggregating photocatalyst materials onto a loose, porous carriers with a large specific surface area can improve catalyst recovery, enhance photocatalyst diversity and utilization rate, and boost photocatalytic efficiency.

4 Cost-effectiveness of photocatalysts preparation

Photocatalytic oxidation technology has broad application prospects in the fields of energy production, pollution treatment, and environmental protection. It is an economical, environmentally-friendly and sustainable way to degrade organic pollutants in wastewater through photocatalytic degradation. However, to promote their wider application, not only does photocatalytic efficiency need to be enhanced through various modification methods, but also the cost of synthetic materials needs to be acceptable. For example, the deposition of Au, Ag, Pd and other noble metals on photocatalytic materials increased the absorption capacity of the catalyst system to visible light^[96-100] but the cost was too high. As shown in Table 7, several photocatalysts were selected, and the cost of photocatalytic materials synthesized by different methods is analyzed based on the market raw material prices and civil electricity charges (0.6 yuan-kWh⁻¹ of electricity) in China.

Through the comparison of several modification methods, it can be concluded that in the preparation process of photocatalytic materials, the high-cost is partly due to electricity costs when using ovens, muffle furnaces and tube furnaces^[89,104,120,126,158], but also the cost of materials^[99-101,103,128-137]. In addition, the method used to synthesize materials is also a major factor in the cost. Given that the sol-gel method to synthesize photocatalytic materials at lower temperatures, the consumption of electric energy is less, but the raw materials used in sol-gel are expensive^[158-163]. Compared to sol-gel methods, hydrothermal synthesis uses cheaper, easily obtained raw materials, but its high-temperature, high-pressure steps increase equipment dependence and constrain its development^[164-168]. In addition, as found in two studies^[169,170], the modification of photocatalytic materials can also be achieved by using functional components in solid waste. This approach simultaneously reduces raw material expenses and achieves waste utilization. Further evaluation of simpler, low-energy synthesis methods is essential for effectively reducing the deployment costs of photocatalytic oxidation processes. Simple synthesis methods offer several key advantages. In terms of ease of implementation, they typically involve fewer steps and require less specialized equipment, making them more accessible to researchers and industries with limited resources. Additionally, simple methods are more user-friendly and can

Table 7 Materials and manufacturing cost of photocatalyst synthesis

Photocatalyst		Cost (yuan)	Ref.
TiO ₂	Cu/TiO ₂ (30 mg)	4.72	[82]
	N/TiO ₂ (50 mg)	1.83	[89]
	Ag/TiO ₂ (25 mg)	1.27	[97]
	SnIn ₄ S ₈ /TiO ₂ (20 mg)	22.76	[109]
	D35 Dye-sensitized/TiO ₂ (8 mg)	11.30	[124]
	Chl Dye-sensitized/TiO ₂ (250 mg)	13.01	[125]
	CQDs/TiO ₂ (4 mg)	18.71	[126]
	NP-Fe ₃ O ₄ /GO-TiO ₂ (80 mg)	19.34	[129]
ZnO	Fe ³⁺ /ZnO (800 mg)	17.34	[85]
	N/ZnO (10 mg)	2.74	[91]
	Ag/ZnO (1 g)	19.30	[98]
	Nd-V-ZnO (180 mg)	14.67	[100]
	Au/Pd/ZnO (0.5 mg)	23.56	[101]
	NaBiS ₂ /ZnO (10 mg)	11.88	[112]
	AC@Au/ZnO (6 mg)	8.16	[141]
g-C ₃ N ₄	N/g-C ₃ N ₄ (50 mg)	7.87	[95]
	Au/g-C ₃ N ₄ (1 g)	18.15	[99]
	CoP/g-C ₃ N ₄ (40 mg)	17.43	[121]
	BPQDs/g-C ₃ N ₄ (30 mg)	21.10	[128]
	ZnO/GO/Ag ₃ PO ₄ (50 mg)	21.22	[137]
	TiO ₂ @g-C ₃ N ₄ @BC (200 mg)	14.32	[158]

be performed without extensive training or highly specialized facilities. For energy efficiency, low-energy synthesis can be achieved by using lower temperatures, shorter reaction times or avoiding energy-intensive steps, such as high-temperature calcination or high-pressure processes. By combining these benefits, simpler and low-energy synthesis methods can significantly lower costs while maintaining effectiveness, making them an attractive option for the widespread adoption of photocatalytic oxidation technologies. Therefore, a comprehensive assessment of raw material costs, energy consumption, waste generation and potential environmental risks is required. Life cycle assessment is helpful for a comprehensive understanding of the environmental impact during the synthesis and use of photocatalytic materials^[171]. This assessment is a systematic approach for assessing the environmental impact of a product or process throughout its entire life cycle, from raw material extraction to final disposal. For example, the preparation of some metal oxide photocatalysts may require calcination at high temperatures,

which consumes a significant amount of energy. By applying life cycle assessment, it is possible to quantify the energy inputs at each stage of the life cycle of the material, from raw material extraction to final product formation.

There are still many problems needing further research for the development of suitable methods for photocatalytic material synthesis. For example, many researches are doing imitative or repetitive work, and do not give attention to innovation of basic theory. Therefore, there is a need to strengthen pioneering and innovative research on basic theories and focus on innovative synthesis methods, such as the use of cheap and readily available materials to synthesize new photocatalytic materials at room temperature.

5 Conclusions and outlooks

Photocatalytic oxidation process has good application prospect

in degrading organic pollutants in wastewater. However, it is still in the experimental stage due to some problems. To apply photocatalytic oxidation technology to the treatment of wastewater from livestock production, the following five key challenges should be prioritized in photocatalytic material modifications and process optimization,

Enhancing visible light utilization. There is a need to expand the visible light response range of photocatalysts and develop new materials. Doping and loading methods can improve photocatalytic materials, but the visible light utilization rate is still low, with limited actual application value. Further research needs to improve visible light utilization ratio.

Sustainable synthesis methods. Compound photocatalysts have complicated synthesis process due to various raw materials and strict reaction conditions like high temperature and pressure. Attention should be given to simplicity, low energy consumption and high efficiency in new material synthesis.

Reducing cost of raw materials. Photocatalysts with noble metal elements increase raw material cost. When developing such materials, improving noble metal utilization rate and adopting convenient synthesis methods are important. Also, low-cost alternatives to noble metals should be considered more, as continuing to develop high-cost noble metal photocatalysts has limited practical value.

Evaluating photocatalysts in practical applications. Evaluating the efficiency of photocatalytic materials using real wastewater and natural sunlight is of practical significance. Current studies often rely on single specific pollutants and simulated light sources, but real wastewater is complex. The presence of multiple pollutants, electrolytes, organic substances and inhibitors (e.g., ammonia and nitrites) in wastewater can reduce photocatalytic efficiency by scavenging radicals, blocking light and competing for active sites. Additionally, natural sunlight is more unstable than simulated light sources, with varying intensity influenced by weather, season and location, which can slow pollutant degradation efficiency.

Exploring photocatalytic reaction mechanisms. The degradation pathways of intermediates and pollutants vary due to the complexity of photocatalytic mechanism, different catalytic materials and environmental factors. Evaluating the

intermediates of photocatalytic reactions is necessary. Raman spectroscopy, HRMS, TOF-MS and DFT calculations and other techniques can be used to study the electronic structure of photocatalysts, predict the energy levels of reactants, intermediates and products as well as the activation energy of reactions, and investigate dynamic processes, such as lattice vibration and hydrogen adsorption in reactions. It is important to be aware that some organic photocatalysts may produce toxic byproducts during degradation processes. Conducting thorough toxicity assessments and ensuring that the materials meet safety standards is essential.

Developing new materials to enhance visible light utilization often involves complex synthesis processes and increased costs, which can constrain scalability and practical application. Meanwhile, the use of noble metals raises material costs, further emphasizing the need for low-cost alternatives and simpler synthesis methods. Understanding the photocatalytic reaction mechanism and intermediate products is crucial for designing better materials with optimized properties, such as improved visible light absorption and charge separation efficiency. However, synthesis complexity material and cost can limit the feasibility of implementing these advanced materials in real-world applications, such as treating actual wastewater with natural sunlight.

Addressing these interconnected challenges requires a balanced strategy that focuses on enhancing visible light utilization, simplifying synthesis processes, reducing costs, and evaluating materials under real-world conditions. This holistic approach is essential for advancing sustainable and practical photocatalyst technologies, ultimately driving their broader application in wastewater treatment. In the short term, the development of photocatalytic materials still needs to focus on enhancing the utilization of visible light and simplifying the synthesis process, so as to promote the application of photocatalytic oxidation technology in actual production. On this basis, further examination of the synthesis and reaction mechanisms of photocatalytic materials to enhance theoretical knowledge reserves. In the long term, the goal is to develop photocatalytic materials and processes that are not only highly efficient but also sustainable and cost-effective. This includes finding low-cost, abundant raw materials and developing energy-efficient synthesis methods that can be easily scaled up for industrial applications.

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Compliance with ethics guidelines

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