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Nonmetal carbon-based photocatalysts for degradation of organic pollutants: a short review

Hongjun Dong[a,b], Xiaoxu Zhang[a], Huihui Wu[a], Chunmei Li[a,*]

[a]*Institute of Green Chemistry and Chemical Technology, School of Chemistry and Chemical Engineering, Jiangsu University, Zhenjiang 212013, P.R. China, emails: lichun_mei_happy@126.com (C. Li), donghongjun6698@aliyun.com (H. Dong), 15500100358@163.com (X. Zhang), 2955178540@qq.com (H. Wu)*
[b]*Key Laboratory of Preparation and Application of Environmental Friendly Materials, Jilin Normal University, Ministry of Education, Changchun 130103, China*

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ABSTRACT

In recent years, semiconductor photocatalysis technique has received more and more attentions for environmental purification due to their unique advantages to solve environmental pollutant problems. To acquire high efficiency, low cost and stable photocatalysts, many nonmetal carbon-based materials have been developed owing to their unique structure and property characteristics. This manuscript presents the recent advance of nonmetal carbon-based photocatalysts for their application in degradation of organic contaminants. We hope it will contribute to deep-level investigation in this field and developing other metal-free semiconductor materials for treating multiple organic pollutants in the future.

Keywords: Nonmetal photocatalysts; Organic pollutants; Degradation; Carbon-based materials

1. Introduction

During the past decades, the continuous increasing environmental pollution problems have become more serious every day owing to a mass of noxious organic pollutants discharging into ecological environment system, which have attracted worldwide attention [1–20]. Many projects have been put forward for removing the pollutants in the environment [21–25]. Some of them have been large-scale applied in wastewater treatment, such as physical/chemical absorption [26,27], membrane separation or filtration [28–30], chemical oxidation [31,32] and so forth. However, the low cost and eco-friendly handling technologies of organic pollutants, especially for being able to tackle persistent organic pollutants with low concentrations, are still greatly needed. Recently, the semiconductor photocatalysis technique has been considered as a potential promising method to deal with the environmental issues because it can directly make the organic pollutants with low concentration and high toxicity decompose completely into non-toxic/low-toxic small molecules/ions under the solar light [33–49]. The involved basic reaction processes are as follows: when semiconductor photocatalyst is exposed to light, it can absorb photons to produce photogenerated electron–hole pairs, after that the electron–hole pairs separate and diffuse to the surface reactivity sites of semiconductor. Following, the photogenerated electrons can be captured by the dissolved oxygen in water to produce superoxide radical ($^{\bullet}O_2^-$). Besides, $^{\bullet}O_2^-$ may further react with H^+ ions in the solution by the reduction of photogenerated electrons to produce (hydroxyl radical) $^{\bullet}OH$. In the meantime, photogenerated holes also may oxidize OH^-/H_2O to produce $^{\bullet}OH$. [9,18,39,50]. As a result, $^{\bullet}O_2^-$, $^{\bullet}OH$ and photogenerated holes as active species with intense oxidization power all can degrade organic pollutants in

* Corresponding authors.

the solution to non-toxic/low-toxic small molecules/ions [40,45,41–56]. The possible carrier transfer behaviors and photocatalytic degradation processes of organic pollutants over the semiconductor photocatalysts under the light excitation are shown in Fig. 1.

As is well known, the high-efficiency photocatalyst as an energy converter plays a most important key role in the above proposed degradation reaction process. At present, some nonmetal materials are usually regarded as a kind of potential promising semiconductor photocatalysts for degradation of various organic pollutants in the environment owing to their attractive and unique features such as multifarious structures, good electrical and thermal conductivity, outstanding stability, low cost, superior stability, rich sources for large-scale production and so forth [52,57–60]. Here we must mention that carbon-based materials, including graphitic carbon nitride (g-C_3N_4) [61–63], graphene oxide (GO) [64–67] and others [68–70] have been developed as a kind of important nonmetal semiconductor photocatalysts, and all of them exhibit outstanding degradation activity and stability for removing the organic pollutants in the environment.

Therefore, in this paper, we will make an overview for the latest advance in relation to the nonmetal carbon-based photocatalysts and their application in photocatalytic degradation of organic pollutants in the solutions. We hope it can provide some new understanding for nonmetal carbon-based photocatalysts in solving the increasingly serious environmental pollution problems caused by organic pollutants.

2. Summarizing overview of carbon-based photocatalysts

Among the reported nonmetal photocatalysts, g-C_3N_4 has been identified as one of the most potential candidates for photocatalytic degradation of organic pollutants in wastewater system due to its unique advantages, such as superior electronic and optical properties, suitable band gap contributing to visible-light absorption, outstanding thermal and chemical stability, low cost, facile synthetic process, etc. [71–74]. However, similar to the reported other kinds of photocatalysts, the bulk g-C_3N_4 usually exhibits relative low

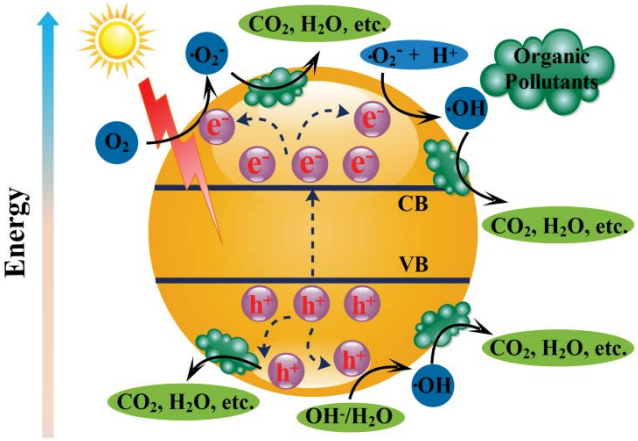

Fig. 1. Possible carrier transfer behaviors and photocatalytic degradation processes of organic pollutants over the semiconductor photocatalyst under the light excitation.

photocatalytic degradation activity because of high recombination probability of photogenerated charge carriers and difficult decomposing organic pollutants completely [75,76]. This is mainly due to the following reasons. On the one hand, the bulk g-C_3N_4 usually has a small specific surface area and fewer surface active sites, resulting in difficult to adsorb pollutant molecules for its degradation to small molecular. On the other hand, the bulk g-C_3N_4 has low potential of valence band, causing its poor oxidizability and g-C_3N_4. Therefore, many methods have been developed to overcome those disadvantages to improve the photocatalytic performance of g-C_3N_4, such as regulating morphologies to increase surface area [77,78], doping heteroatoms to adjust electronic band structure [79], and constructing heterostructures by coupling with other metal-free materials to facilitate separation of photogenerated charge carriers [80].

2.1. Morphological control of g-C_3N_4

The morphological control is one of the earliest means to improve the photocatalytic activity of g-C_3N_4, For instance, ultrathin and high-yield two-dimension (2D) g-C_3N_4 nanosheets with a thickness of 2 nm were prepared by means of calcining bulk g-C_3N_4 under H_2 air flow [81]. It revealed that the enhanced photocatalytic activity for dye degradation compared with that bulk g-C_3N_4 originates from its high surface area and the low recombination possibility of photogenerated charge carriers. Besides above 2D structure, constructing three dimensional (3D) multiaperture architectures is also an attractive option to boost photocatalytic degradation performance of g-C_3N_4. Using silica nanoparticles as templates, Cui et al. [82] synthesized mesoporous g-C_3N_4 by a facile thermal-induced polymerization of NH_4SC. The photocatalytic experimental results displayed that the achieved mesoporous g-C_3N_4 possessed the higher activity than that of nonporous g-C_3N_4 for degradation of chlorophenol and phenol in the solution under the visible light. It is because that the unique porous structure increases the surface area and light harvesting capacity, which contribute to the generation of active oxy-radicals in water. Moreover, g-C_3N_4 with 3D ordered macroporous (3DOM) structure was constructed through a facile thermal condensation under the assistance of colloidal crystal as template [83]. The as-prepared 3DOM g-C_3N_4 photocatalyst exhibited the superior degradation activity for dye and the removal rate nearly reached up to 100% within 40 min under visible-light irradiation, which was attributed to the ordered 3D macroporous structure that facilitates light absorption, reactant diffusion and charge separation. In addition, Bai et al. [84] group also realized the transformation of g-C_3N_4 from nanoplates to nanorods by a facile reflux technique in the mixed solvents of CH_3OH and H_2O without using any templates (Fig. 2a). The morphology evolution mechanism of g-C_3N_4 from nanoplates to nanorods was that nanoplates may go through exfoliation, regrowth and then a lamellar structure rolling process. As shown in Fig. 2b, the resulting photocatalytic activity of g-C_3N_4 nanorods was higher than that of nanoplates for dye degradation under visible light, which may be due to the increase of active facets and elimination of surface defects during the reflux process. Furthermore, the other various

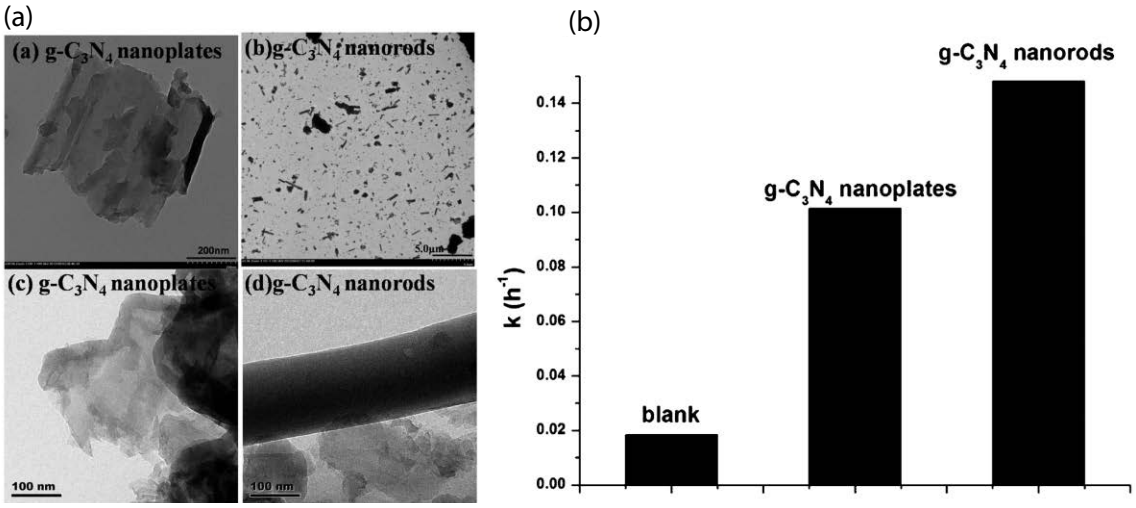

Fig. 2. TEM images of g-C_3N_4 nanoplates and g-C_3N_4 nanorods (a); apparent rate constants for MB photodegradation over g-C_3N_4 nanoplates and g-C_3N_4 nanorods under visible light and (b). Reprinted with permission from the study of Bai et al. [84]. Copyright © 2013, American Chemical Society.

morphological control of g-C_3N_4 have also been performed by virtue of some innovative synthetic strategies, such as successfully synthesized nanofibers [85], quantum dots [86], micro strings [87], tubular [88], etc. Most of them exhibited outstanding photocatalytic performances for degrading diversified organic pollutants, including dyes, antibiotics, phenol and so forth.

2.2. Doping modification of nonmetal elements into g-C_3N_4

Doping foreign nonmetal elements into g-C_3N_4 has also been proved to be an efficiency strategy to control the position, structure and composition of energy band for improving photocatalytic activity [89–96]. For instance, boron-doped g-C_3N_4 photocatalyst has also been prepared by using the melamine and boron oxide as precursors, which apparently enhanced the photocatalytic activity for degrading RhB dye owing to the improvement of light absorption and adsorption capability [79]. Additionally, the fluorinated g-C_3N_4 (CNF) heterogeneous photocatalyst was synthesized using ammonium fluoride as a cheap fluorine source [94]. As revealed by UV–Vis spectrum (Fig. 3a), the optical band gap and the semiconductor properties of the CNF have indeed slightly been changed owing to fluorination, with extension of the visible light response and decrease of band gap. Therefore, the photocatalytic activity was improved for the oxidization reaction of benzene under the visible light (Fig. 3b). Simultaneously, phosphate-doped g-C_3N_4 nanomaterial has been used as a valid photocatalyst for colorless pollutants degradation [97]. The photocatalytic activity of g-C_3N_4 was obviously improved after modification with phosphoric acid for degrading gas-phase acetaldehyde and liquid-phase phenol, which resulted from increase in adsorbed O_2 molecule which not only prolongs the lifetime but also enhances the separation of photogenerated charge carriers. Up to now, based on engineering of electronic

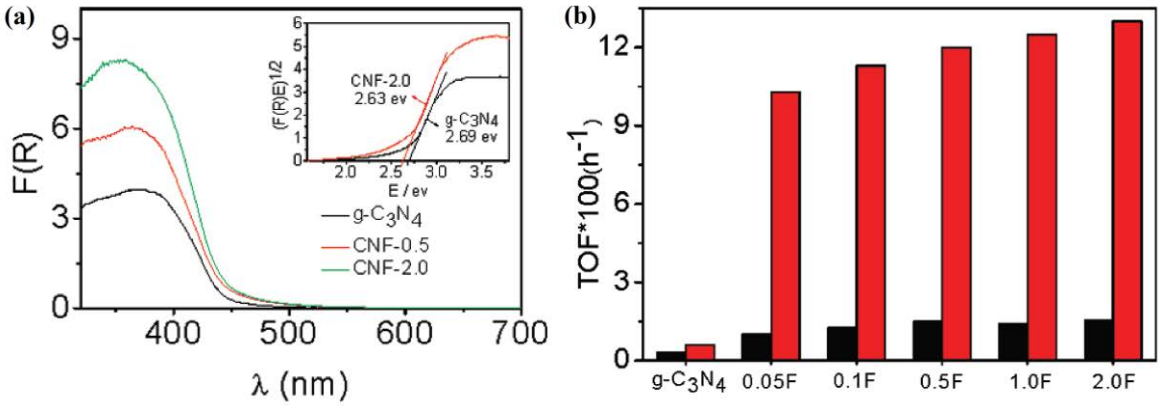

Fig. 3. UV–Vis DRS spectra (a) and optical band gap (inset) of g-C_3N_4 and CNF-*x*; catalytic oxidation of benzene using CNF-*x* (b). Reprinted with permission from the study of Wang et al. [94]. Copyright© 2010, American Chemical Society.

structure, an increasing number of g-C_3N_4 doped with the nonmetal such as B, C, N, S, O, P, F, Br, I, etc have been investigated in succession for enhancing the photocatalytic performance [89–97].

2.3. Constructing heterostructure of g-C_3N_4 with other nonmetal materials

Constructing heterostructure with other functional nonmetal components was an effective way to improve the photocatalytic activity of g-C_3N_4 for degrading organic pollutants. For instance, Zhang et al. [98] synthesized a carbon dots decorated g-C_3N_4 photocatalyst by means of impregnation-thermal strategy. The results demonstrated that this composite heterostructure with low carbon dots content showed promising photocatalytic activity for phenol degradation under the visible light. This can be derived from the extended visible light absorption range induced by the up-converted photoluminescence feature of carbon dots and the efficient electron–hole separation resulted from the suitable band alignment in the composite structure. Meanwhile, Pitre et al. [69] synthesized N-doped carbon dots (NCDs) modified g-C_3N_4 composite heterostructure via a facile polymerized method. As-prepared NCDs/g-C_3N_4 heterostructure was constructed through loading NCDs nanoparticles onto the interlayers and surface of g-C_3N_4 via π–π stacking interactions. The photocatalytic activity toward degradation of indomethacin was apparently higher than that of g-C_3N_4 and CDs/g-C_3N_4 under the visible light irradiation, which ascribed the unique up-converted photoluminescence property, narrowing band gap and efficient charge separation. 3D porous g-C_3N_4/GO aerogel (CNGA) has also been constructed by the hydrothermal co-assembly scheme of g-C_3N_4 and GO nanosheets [99], which exhibited much higher removal methyl orange activity than that of 2D hybrid counterpart, pure g-C_3N_4 and the most of representative g-C_3N_4-based photocatalysts because the high interconnected porous network rendered numerous pathways, strong adsorption and multi-reflection of incident light (Fig. 4). Other nonmetal g-C_3N_4-based composite materials have also been prepared by several strategies for degradation of organic contaminants, such as PANI/g-C_3N_4 [100], polyimide/g-C_3N_4 [80], carbon dots/g-C_3N_4 [101], GO/g-C_3N_4 [102] and so on.

2.4. Graphene and its derivatives

Graphene is another kind of frequently used inorganic material as a functional component of nonmetal photocatalysts to promote the degradation activity for removing organic pollutants because it can not only increase the adsorption capability but also be served as an electron-transfer medium to facilitate photogenerated charge separation [103–108]. For example, Zhang et al. [109] reported the synthesis of 3D carbon nanotubes-pillared graphene oxide (GO) or reduced graphene oxide (rGO) composite photocatalysts by chemical vapor deposition strategy, which resulted in perfect photocatalytic activity in degradation of typical RhB dye (Fig. 5). The synergistic effect between 1D carbon nanotubes and 2D rGO nanosheets reduced the dynamic resistance transport and recombination probability of charge carriers effectively, which was a crucial reason for enhancing photocatalytic degradation activity. In recent years, high monodispersed silica nanocrystals were orderly grown on the single-layer graphene sheet in the modified Stöber process by using vinyltriethoxysilane and GO as the precursors, in which GO is concurrently reduced to rGO by ammonia. The silica nanocrystals were exclusively and homogenously distributed on rGO and had excellent crystallinity, which derived from the chemical similarity and synchronized reaction between vinyl-silanol and GO. The prepared composite exhibited superior photocatalytic degradation activity for RhB and 2,4-dichlorophenol relative to isolate and physically mixed graphene and SiO_2 [110]. Furthermore, the benzothiadiazole (BTZ)-based conjugated porous polymer (CMP) loaded into graphene aerogel has been reported, which is prepared by a

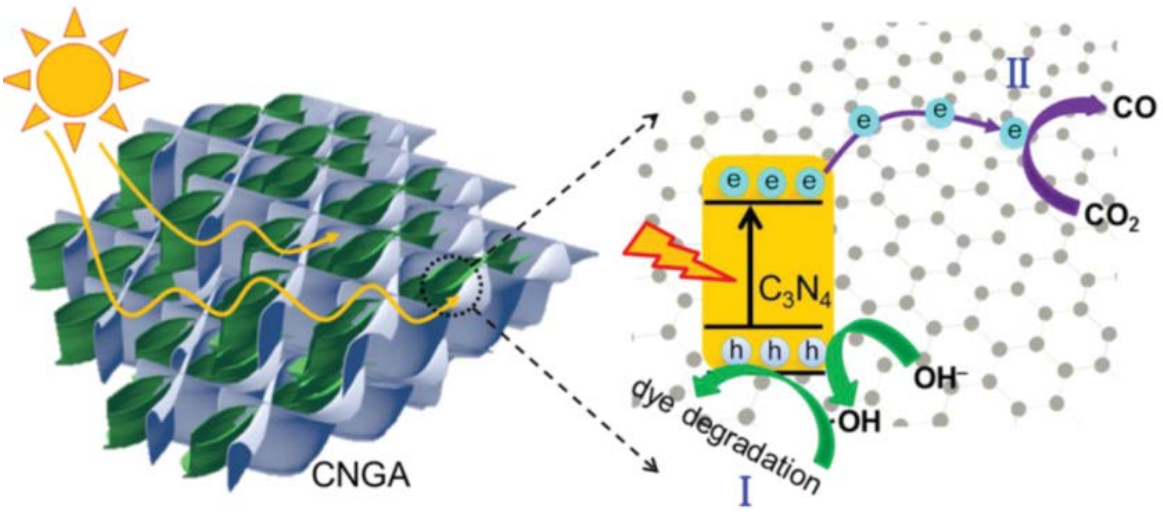

Fig. 4. Schematic diagram for illustrating the photodegradation (I) and photoreduction (Π) processes over CNGA under visible-light irradiation. Reprinted with permission from the study of Fang et al. [101]. Copyright© 2015, American Chemical Society.

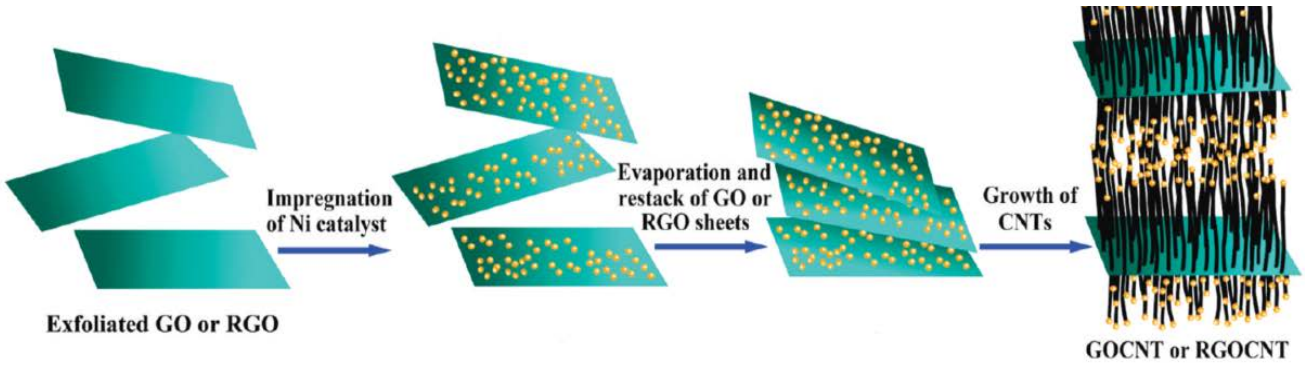

Fig. 5. Experimental steps of pillaring GO and RGO platelets with CNTs. Reprinted with permission from the study of Tsang et al. [111]. Copyright© 2010, American Chemical Society.

one-step green hydrothermal reaction process between 2D GO and the CMP under mild conditions. It exhibits photodecomposition of MO ability under visible light. This result explains that the CMP-loaded GA can act as an efficient metal-free material for photocatalytic application [111]. As effective electron acceptor, graphene-based composite photocatalysis have also been reported 2 years ago. For instance, Gong et al. [112] prepared GO enwrapped polyimide (PI) composite photocatalyst by the simple ultrasonic chemical process, which showed the stable metal-free photocatalyst for 2,4-dichlorophenol degradation under visible light. After loading electron acceptor GO, the GO/PI composites displayed enhanced photocatalytic performance obviously and it was approximately 4.5 times as high as that of bare PI. This results is due to the efficient injection of photogenerated electrons from PI to GO, and promotes the separation of charge carrier [112].

In addition, Chen et al. [113] introduced porphyrin nanoparticles into macroscopic rGO by a facile vacuum filtration method of the co-colloids of GO and porphyrin nanoparticles followed by gaseous reduction. The as-prepared composite photocatalyst showed enhanced photocatalytic activity for degrading RhB and MB dyes under the visible light, and exhibited excellent stability and easy recovery compared with each hybrid monomer. This is because that the large contact interface between rGO and porphyrin nanoparticles was conducive to the strong interaction and boosted electron–hole pair separation during the photocatalytic reaction process. As a result, the obtained composite may act as a potential photocatalytic material for environmental remediation in the future. Other graphene-based composite photocatalysts, such as polyaniline/rGO [114], SiC/graphene [115], etc. also confirmed graphene had promotional effect on the photocatalytic degradation reaction of various organic pollutants. Besides, graphene can also act as photosensitive materials for enhanced photocatalytic degradation activity. However, most of the above photocatalysts involves metallic semiconductor, it does not merit any additional discussion here.

Moreover, elements doping in graphene was also an effective method for improving photocatalytic activity. Tang et al. [116] prepared boron-doped rGO photocatalyst by means of one-step reflux route, which showed greatly improved photocatalytic activity toward the degradation of RhB dye in contrast to the non-doped rGO under the visible light. It can be due to the more efficient electron transfer from the excited state of RhB dye to boron-doped rGO with superior electron transfer capacity and electronic conductivity. The controlled experiments also proved RhB dye degradation mainly originated from the photosensitization rather than photoexcitation process in band gap of the prepared sample.

2.5. Other nonmetal photocatalysts and its derivatives

Besides the above frequently used nanomaterials, some other nonmetal photocatalysts had been applied effectively to photocatalytic degradation of organic pollutants. For instance, using a simple ball milling method, this group also exploited black-red phosphorus heterostructure and its photocatalytic activity was comparable with CdS [117]. Zhang et al. [55] prepared resorcinol–formaldehyde resin polymers, the suitable band gap energy of which resulted in its good visible light response. This resin can efficiently oxidize organic pollutants and the photocatalytic activity was further markedly enhanced after loading rGO [55]. In this year, the GR-BP hybrid photocatalysts was synthesized by one-pot chemical vapor transport approach, in which the P atom was successfully incorporated into GR through the formation of P-C bond (Fig. 6) [118]. Remarkably, this photocatalyst represented high photocatalytic activity for 2-chlorophenol degradation, which is due to the higher carrier separation efficiency caused by the direct band gap of BP and carrier mobility of GR. Besides, the little increasing of P-O bond was observed after GR-BP photocatalysts being exposed in air for 15 d, which means the BP stability was enhanced significantly. Furthermore, carbon nanotubes with highly defective (DF-CNTs) were prepared using a heat-treatment technique, which exhibits photocatalytic degradation H_2O_2 ability in the range of visible light. The as-prepared DF-CNTs present the relatively high surface energy causing local lattice reordering and intertube reorientation, which lead to the formation of more topological defects. As a result, light absorption capacity takes shape and thus induced electron/hole pair's generation over the surface of DF-CNTs [119]. Furthermore, phosphorus fibers [120] had also been confirmed as effective photocatalysts for degrading organic pollutants. The summary of

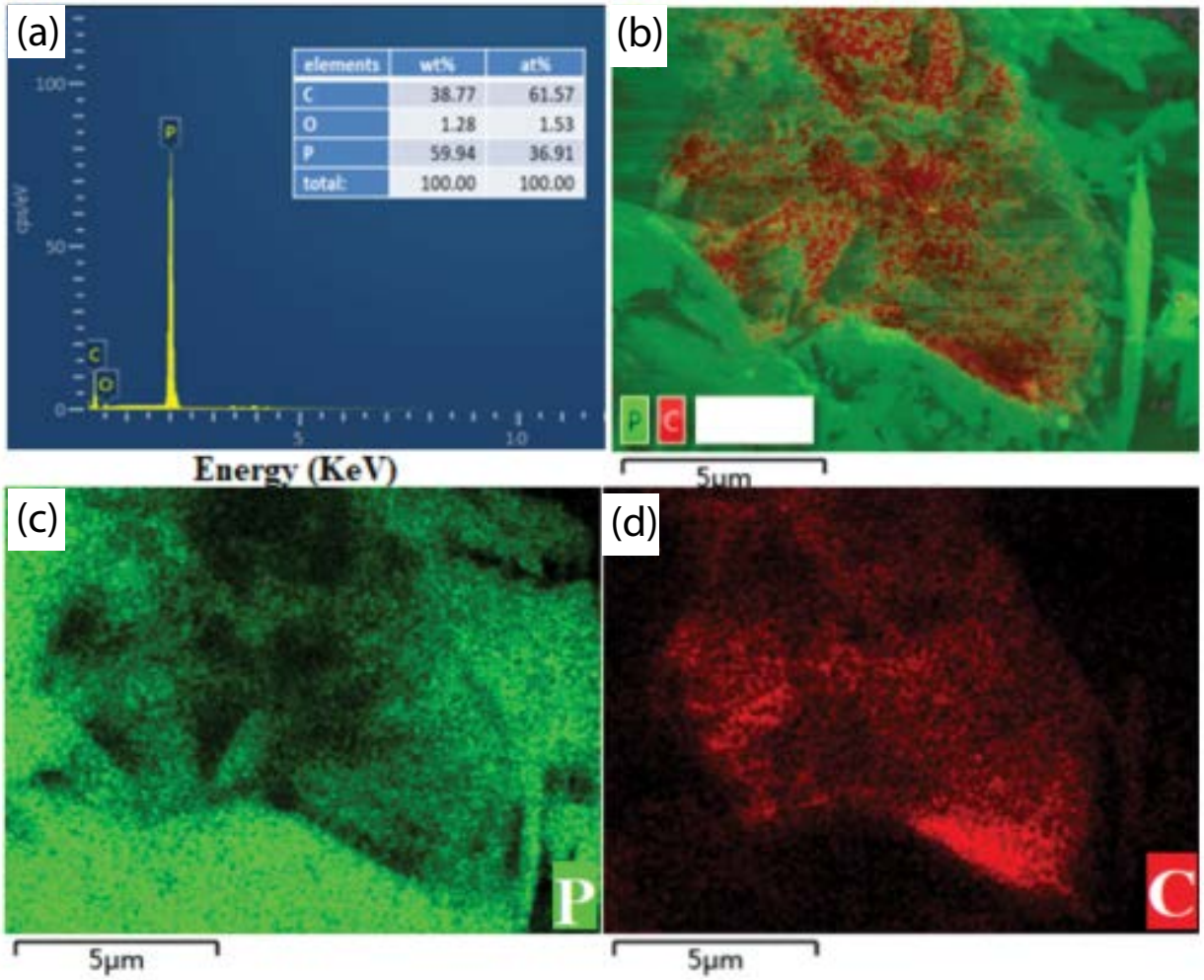

Fig. 6. Images of the synthesized GR-BP hybrid: (a) EDS spectrum of 30% GR-BP hybrid, (b) EDS elemental dot-mapping, (c) P element dot-mapping and (d) C element dot mapping. Reprinted with permission from the study of Shen et al. [120]. Copyright©2019, Elsevier Publishing Group.

Table 1
Photocatalytic degradation of organic pollutants over some nonmetal photocatalysts

Photocatalysts (mg)	Pollutants (mg/L)	Light sources (W)	λ (nm)	Degradation rates (min)	References
Porous g-C_3N_4 (25)	MB (10)	Xe lamp (500)	≥420 nm	70% (150)	[121]
3D Macroporous g-C_3N_4 (70)	RhB (10)	Xe lamp (300)	≥420 nm	100% (40)	[83]
Mesoporous g-C_3N_4 (40)	RhB, 4-CP (1.2×10^{-4} M)	Xe lamp (300)	≥420 nm	100% (60), 96% (90)	[82]
Ultrathin g-C_3N_4 (80)	MB (10)	Xe lamp (500)	Visible light	60% (180)	[81]
P-doped g-C_3N_4 (100)	Phenol (10)	Xe lamp (150)	–	80% (60)	[97]
B-doped g-C_3N_4 (200)	RhB, MO (4)	Xe lamp (300)	≥420 nm	100% (40), 100% (300)	[79]
C-dots/g-C_3N_4, (50)	Phenol (10)	Xe lamp (300)	≥400 nm	100% (200)	[98]
PANI/g-C_3N_4 (100)	MB (10)	Xe lamp (500)	≥420 nm	92.8% (120)	[101]
GO/g-C_3N_4 (---)	RhB, 2,4-DCP (10)	Xe lamp (100)	≥400 nm	94.2% (155), 87.1% (240)	[67]
B-doped rGO (10)	RhB (0.01M)	Xe lamp (300)	≥420 nm	100% (130)	[116]
PAN1/GO (10)	MO, MB, RhB (10)	Natural sunlight	–	100% (40, 140, 100)	[114]
Black–red phosphorus (50)	RhB (10 ppm)	Xe lamp (300)	≥420 nm	95% (120)	[117]
Carbon nanotubes (1.5 mL)	H_2O_2 (10 mM)	W lamp (500)	–	90% (180)	[119]
Phosphorus fibers (2)	RhB (10 ppm)	Xe lamp (300)	≥420 nm	46.4% (360)	[120]

photocatalytic degradation activity, pollutant type and reaction conditions of most metal-free photocatalysts are shown in Table 1.

3. Conclusions and perspectives

In summary, this review summarized the recent advance involving nonmetal photocatalysts for degradation of organic pollutants in environmental remediation. Up to now, a series of metal-free photocatalytic materials including g-C_3N_4, graphene, graphene derivatives and other polymer-based semiconductor materials have been confirmed to possess potential application in removing organic pollutants. g-C_3N_4 is the representative nonmetal photocatalyst for degrading and mineralizing various organic pollutants effectively under the visible light. Further optimizing parameters in degradation process based on g-C_3N_4 is still crucial in the photocatalytic oxidation processes, which can obviously influence the degradation efficiency of pollutants in water. Despite the obvious progress has been made, it is still challenging to investigate other important and significant topics, including the relationship between electronic properties and defects, the surface active sites and photodegradation property of the metal-free photocatalysts. Additionally, it is also worth investigated on a few topics related to practical application based on metal-free photocatalysts, such as photocatalytic CO_2 reduction [122], photocatalytic water splitting for H_2 evolution [123], photocatalytic removal of NO_x [52], photoreduction of Cr(VI) [124] and so on. Admittedly, we all knew that the development of nonmetal photocatalysts is continuously evolving for practical applications in environment purification. Although some important progress has been acquired in this field at present, we still need further efforts for exploiting the large-scale utilization of nonmetal photocatalysts to achieve the goal of environmental purification.

Acknowledgements

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