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Review

Graphitic carbon nitride based nanocomposites for the photocatalysis of organic contaminants under visible irradiation: Progress, limitations and future directions

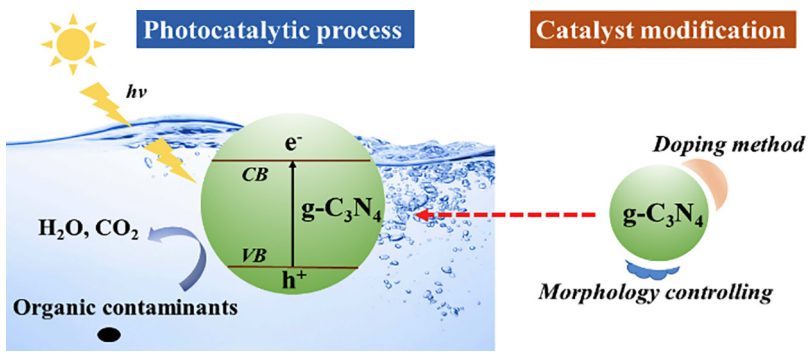
Bentuo Xu [a,b], Mohammad Boshir Ahmed [a], John L. Zhou [a,*], Ali Altaee [a], Gang Xu [b], Minghong Wu [b]

[a] School of Civil and Environmental Engineering, University of Technology Sydney, 15 Broadway, NSW 2007, Australia
[b] School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, China

HIGHLIGHTS

- g-C_3N_4 catalyst performs better than TiO_2 in visible photocatalysis of dyes.
- Photodegradation by g-C_3N_4 is particularly enhanced by K-doping.
- Compound doping especially Ag_3PO_4-g-C_3N_4 improves photodegradation effect.
- Co-doping can degrade multiple contaminants with high degradation rate constant.
- Synthesized g-C_3N_4 nanocomposites have shown good reusability.

GRAPHICAL ABSTRACT



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ABSTRACT

Graphitic carbon nitride (g-C_3N_4) has drawn great attention recently because of its visible light response, suitable energy band gap, good redox ability, and metal-free nature. g-C_3N_4 can absorb visible light directly, therefore has better photocatalytic ability under solar irradiation and is more energy-efficient than TiO_2. However, pure g-C_3N_4 still has the drawbacks of insufficient light absorption, small surface area and fast recombination of photogenerated electron and hole pairs. This review summarizes the recent progress in the development of g-C_3N_4 nanocomposites to photodegrade organic contaminants in water. Element doping especially by potassium has been reported to be an efficient method to promote the degradation efficacy. In addition, compound doping improves photodegradation performance of g-C_3N_4, especially Ag_3PO_4-g-C_3N_4 which can completely degrade 10 mg L^{-1} of methyl orange under visible light irradiation in 5 min, with the rate constant (k) as high as 0.236 min^{-1}. Moreover, co-doping enhances the photodegradation rate of multiple contaminants while immobilization significantly improves catalyst stability. Most of g-C_3N_4 composites possess high reusability enabling their practical applications in wastewater treatment. Furthermore, environmental conditions such as solution pH, reaction temperature, dissolved oxygen, and dissolved organic matter all have important effects on the photocatalytic ability of g-C_3N_4 photocatalyst. Future work should focus on the synthesis of innovative g-C_3N_4 nanocomposites for the efficient removal of organic contaminants in water and wastewater.

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* Corresponding author.
E-mail address: junliang.zhou@uts.edu.au (J.L. Zhou).

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1. Introduction

As the world is facing increasing challenges in the field of energy crisis and environmental pollution, the use of renewable energy to control environmental pollution is of high priority. As an inexhaustible and environmental friendly resource, solar energy is considered the most ideal power, which enables photodegradation to be a favourable technology for controlling environmental contamination (Ahmed et al., 2017; Bahnemann, 2004; Malato et al., 2002). By comparison, other treatment methods such as electrochemistry (Stilwell and Park, 1988), filtration (Simon et al., 2009), biodegradation (Pagga and Brown, 1986), and membrane bioreactor (Quintana et al., 2005) all have the drawback of energy consumption. However, for the practical applications of photolysis, low efficacy of solar energy utilization is the main obstacle. Thus, photocatalysis, especially semiconductor photocatalysis, becomes popular due to the advantages of using renewable resources, cost-effectiveness, safety, and comparatively high efficacy of pollutant removal (Xu et al., 2017a). For example, titanium dioxide (TiO_2), zinc oxide (ZnO), bismuth sulfide (Bi_2S_3), and carbon nitride (C_3N_4) all have been reported to perform well in photocatalysis (He, 2017; Konyar et al., 2017; Zhang et al., 2007; Zhou et al., 2017). These semiconductors possess the ability to adsorb the light and produce photogenerated electron and hole pairs, which have redox ability to degrade environmental contaminants. In addition, the doping of catalyst such as TiO_2 by gold (Au) has enabled the catalyst to perform significantly better under visible light (Sornalingam et al., 2018).

Notably, C_3N_4 is a non-toxic, abundant and low cost material, and has a metal-free property compared with other common catalysts (Zhao et al., 2015b). Teter and Hemley (1996) conducted the first research on the C_3N_4 material and provided five molecular structures: β-C_3N_4, α-C_3N_4, g-C_3N_4, p-C_3N_4, and c-C_3N_4 (Fig. S1). Among them, g-C_3N_4 is most stable at room temperature and atmospheric pressure with the lowest density. From the end of the twentieth century, g-C_3N_4 has been extensively applied in many fields, including fuel cells, photocatalysis, gas storage, reduction of carbon dioxide, and water splitting to produce hydrogen (Shi et al., 2017; Wen et al., 2017; Zhu et al., 2017). As a catalyst, g-C_3N_4 has the band gap energy of 2.65 eV which enables it to directly absorb visible light in the 400–800 nm region (Chen et al., 2015b; Patnaik et al., 2016). Moreover, the redox ability of the transferred photoexcited carriers and excellent chemical stability make it potentially suitable for the solar energy conversion and pollutants degradation. By comparison, TiO_2, as a traditional catalyst, owns higher band gap energy of 3.2 eV than g-C_3N_4 and thus only UV light (wavelength below approximately 360 nm) can be utilized in the TiO_2 catalytic system, indicating that it is not energy efficient (Xu et al., 2017a). Although, pristine g-C_3N_4 still has three main disadvantages including insufficient absorption of light energy, low surface area and fast recombination of photogenerated electron-hole pairs, leading to a comparatively low photodegradation efficacy (Cao et al., 2015; Mamba and Mishra, 2016). Therefore, many routes have been developed to solve such problems and promote the photocatalytic activity. For example, metal doping, non-metal doping, and coupling with other semiconductors are known to enhance the effectiveness of the catalyst. The fusion of two π-conjugated systems, such as g-C_3N_4 combining with BiOBr system as shown in Fig. 1A, not only enhances the charge separation of electron and hole from the single material, but also further increases the usable region of the solar spectra (Patnaik et al., 2016). Moreover, morphology controlling is another effective method to promote the degradation efficacy as it could significantly enlarge the surface area of the catalyst, thereby its adsorption ability of both light energy and contaminants is promoted (Niu et al., 2012).

g-C_3N_4 and modified g-C_3N_4 composites have been successfully applied to the remediation of organic contaminants. For example, dyes such as rhodamine B (RhB), methyl orange (MO), basic fuchsin (BF) and methylene blue (MB) were treated using synthesized g-C_3N_4 composites (Liao et al., 2012; Liu et al., 2011; Wang et al., 2014; Yan et al., 2009), with better degradation efficacies than by TiO_2 and other types of semiconductor. Using MO photocatalysis as an example, pure g-C_3N_4 degraded 15% MO within 20 min, compared with <10% MO removal by TiO_2. Furthermore, g-C_3N_4 doped with CdS could degrade 90% MO under the same situation (Fu et al., 2013). In comparison, the catalyst ZnO/SnO_2 photodegraded approximately 50% MO within 20 min while Bi_2S_3 nanorods just removed about 10% MO under similar conditions (Wang et al., 2004; Wu et al., 2010). Dye compounds are not only widely used in the textile, printing and plastic industries, but also hard to be degraded in water due to their complex composition, deep color, and chemical stability. Dyes represent some of the principal contaminants in the wastewater from the textile industry and can absorb visible light irradiation (Wu et al., 1998). Other organic compounds such as phenol, atrazine, and humic acid have also been reported to be treated with g-C_3N_4 composites in photocatalysis system, indicating that g-C_3N_4 has the photocatalytic ability to degrade different kinds of organic contaminants from water and wastewater (Jo et al., 2016; Ma et al., 2016b; Ren et al., 2014).

Up to now there have been several publications on the application of g-C_3N_4 in water splitting, H_2 production, CO_2 reduction and

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