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Developing polyetherimide/graphitic carbon nitride floating photocatalyst with good photodegradation performance of methyl orange under light irradiation

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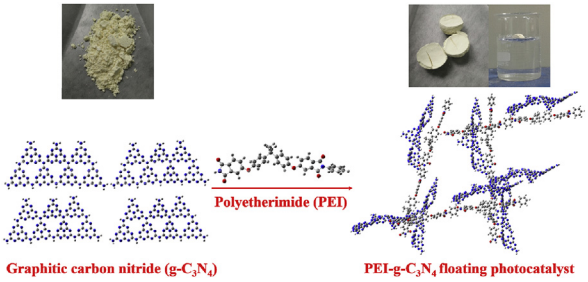
HIGHLIGHTS

- Polyetherimide-graphitic carbon nitride (PEI-g-C_3N_4) floating photocatalyst is synthesized by using PEI to bind g-C_3N_4 together.
- The photodegradation performance of PEI-g-C_3N_4 is from the contribution of g-C_3N_4 and PEI just works as linker.
- PEI-g-C_3N_4 can break both N=N bond and aromatic ring in methyl orange (MO), showing that it has strong photo-oxidation ability.
- The photodegradation efficiency of MO by PEI-g-C_3N_4 is higher than that by g-C_3N_4 without stirring.

GRAPHICAL ABSTRACT



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ABSTRACT

Polyetherimide-graphitic carbon nitride (PEI-g-C_3N_4) floating photocatalyst has been synthesized by using polyetherimide (PEI) as linker to bind graphitic carbon nitride (g-C_3N_4) together. XRD and XPS analysis for PEI-g-C_3N_4 show that the interaction between PEI and g-C_3N_4 does not disturb the structure of g-C_3N_4. FTIR, TEM and theoretical results suggest that the long chain PEI binds g-C_3N_4 particles together to form PEI-g-C_3N_4 via hydrogen bonding interaction. Based on photodegradation results of methyl orange (MO), PEI can not photodegrade MO and just works as linker in PEI-g-C_3N_4, while the photodegradation performance of PEI-g-C_3N_4 is from the contribution of g-C_3N_4. Total organic carbon (TOC) analysis show that nearly 47% organic carbon has been converted into inorganic carbon after photodegradation, suggesting that PEI-g-C_3N_4 can destroy both N=N bond and aromatic rings in MO under light irradiation. The photodegradation efficiency (91%) of MO by g-C_3N_4 is higher than that (80%) by PEI-g-C_3N_4 with stirring. But, the photodegradation efficiency (37%) of MO by g-C_3N_4 is lower than that (55%) by PEI-g-C_3N_4 without stirring. This is the advantage of floating photocatalyst with respect to the powder photocatalyst since the former can utilize more solar energy than the latter when stirring is not available.

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Abbreviation

MO	methyl orange
PEI	polyetherimide
g-C_3N_4	graphitic carbon nitride
PEI-g-C_3N_4	polyetherimide-graphitic carbon nitride, in which the used amount of polytherimide is 45 mg
PEI(15)-g-C_3N_4	polyetherimide-graphitic carbon nitride, in which the used amount of polytherimide is 15 mg
PEI(75)-g-C_3N_4	polyetherimide-graphitic carbon nitride, in which the used amount of polytherimide is 75 mg
TOC	total organic carbon

1. Introduction

The energy from sun to earth in 1 h is 4.3×10^{20} J (Arakawa et al., 2001), which makes that solar energy become one of the attractive new energies to replace the traditional fossil energy. Photocatalytic materials can degrade pollutants under light irradiation, and photocatalysis has been considered as the one of these promising advanced oxidation technologies to treat environmental pollutants in air (Kibanova et al., 2009) and water (Malati, 1995). So far, lots of photocatalysts have been developed to harvest solar energy, mainly including inorganic materials (e.g. metal oxides, nitrides, sulfides, phosphides) (Kudo and Miseki, 2009), organic metal complex (e.g. Ru-complex, Pt-complex) (Ravelli et al., 2009). However, it has the risk of metal dissolution when these photocatalysts containing metal elements are used to degrade the environmental pollutants in water bodies (Han et al., 2010). Graphitic carbon nitride (Wang et al., 2012) and carbon-doped boron nitride (Huang et al., 2015) are polymeric photocatalysts, which are synthesized under high temperature through polycondensation reaction and have good photocatalytic performance under visible light irradiation. They just have C, N, H, B elements, as well as are stable under acidic and basic conditions. Thus, it has not the risk of nitrogen loss when these metal-free polymeric photocatalysts are used to degrade the environmental pollutants in water bodies. Until now, many efforts have been done to promote the application of these polymeric photocatalysts in the purification of the contaminated water bodies (Wang et al., 2013; Guo et al., 2015).

At the present time, photocatalysis technology has been used to degrade air contaminants in a large scale. For example, photocatalytic coating has been used to remove the formaldehyde in the room (Bourgeois et al., 2012). Photocatalytic cement are applied to degrade the NO_X gas released by cars (Bo et al., 2014). Self-cleaning glass also adopts the photocatalytic technology to covert these lipophilic contaminants into hydrophilic molecules by doping photocatalytic material into the glass (Cedillo-González et al., 2014). However, the application of photocatalytic technology in the purification of the contaminated water under natural condition are still not too much. This is main due to the following reason: most of the reported photocatalysts are powder and stirring is indispensable during photodegradation process. These photocatalysts will sink into the bottom of water bodies and can not effective utilize solar energy if stirring lacks. In addition, most of the reported photocatalysts just have strong photodegradation performance under the irradiation of these powerful light source (such as xenon lamp and high-pressure mercury lamp) (Ghosh et al., 2015; Marchena et al., 2016), which further increase the cost using photocatalysts to degrade contaminants in water bodies. For promoting the application of the photocatalytic technology in the treatment of the contaminated water, some floating photocatalyst have been synthesized so far. For example, TiO_2 and B–N–TiO_2 have been combined with expanded perlite together to form the floating photocatalysts for the degradation of organic pollutants (Dlugosz et al., 2014; Xue et al., 2016).

Based on other and our previous reports (Liu et al., 2011; Lin and Wang, 2014; Guo et al., 2013), graphitic carbon nitride (g-C_3N_4) photocatalyst has good photodegradation performance for organic pollutants, such as methyl orange and methylene blue. Many effort has been devoted to promote the application of g-C_3N_4 in energy and environment fields. For example, B (Yan et al., 2010), F (Wang et al., 2010), P (Zhang et al., 2010) and S (Liu et al., 2010) elements have been doped into the g-C_3N_4 skeleton to improve its photocatalytic activity. Sensitization (Guo et al., 2012) and heterojunction (Di et al., 2010) strategies have also been used to increase the photocatalytic activity of g-C_3N_4. However, g–C_3N_4–based floating photocatalysts has not been reported so far. Polyetherimide (PEI) is one kind of polymer with high stability and viscosity, which has been wide used to glue materials (such as zeolite and graphene) together to form membrane or foam (Husain and Koros, 2007; Ling et al., 2013).

Herein, polyetherimide-graphitic carbon nitride floating photocatalyst (PEI-*g*-C_3N_4) has been synthesized by using PEI as linker to bind g-C_3N_4 together. The objectives of this work are included as the following: (1) investigate the interaction between PEI and g-C_3N_4 as well as the formation mechanism of PEI-g-C_3N_4; (2) investigate the photocatalytic performance of PEI-g-C_3N_4 under light irradiation of table lamp by using methyl orange as target pollutant.

2. Experimental and theoretical methods

2.1. Regents

Urea, polyetherimide (PEI), dichloromethane and methyl orange (MO) were all purchased from the Sinopharm Chemical Reagent limited corporation, P.R. China. Polyetherimide is a transparent and yellow particle (Fig. S1).

2.2. Preparation of g-C_3N_4

g-C_3N_4 was prepared according to the following way: 40 g urea powder was put into an alumina crucible with a cover, and dried in oven at 80 °C for 2 h. Then, the alumina crucible containing 40 g urea with a cover was put into a muffle furnace and heated at 520 °C for 1 h (Liu et al., 2011). Heating rate was 7 °C/min. The obtained sample was yellow powder (Fig. S2).

2.3. Preparation of polyetherimide-graphitic carbon nitride (PEI-g-C_3N_4) floating photocatalyst

5 ml dichloromethane was put into a 10 ml centrifuge tube, followed by the addition of three PEI particles (about 45 mg) into the centrifuge tube. The concentration of the PEI solution is 9 mg/ml. Then, ultrasonication was used to promote the dissolution of PEI in dichloromethane to acquire PEI solution. After that, 30 mg g-C_3N_4 powder was put into the PEI solution in centrifuge tube with the subsequent ultrasonication and shake for 30 min. Finally, PEI-g-C_3N_4 suspension was centrifuged for 10 min with the rotating speed of 4000. The centrifugate was dried at room temperature since dichloromethane was easily volatile. The obtained solid sample through centrifugation had a wafer shape and was able to float on water (please see Fig. S3). This sample was named as PEI-g-C_3N_4