



Synthesis of magnetic CoFe₂O₄/g-C₃N₄ composite and its enhancement of photocatalytic ability under visible-light



Shuquan Huang^a, Yuanguo Xu^a, Meng Xie^b, Hui Xu^a, Minqiang He^{a,*}, Jiexiang Xia^a, Liying Huang^a, Huaming Li^{a,**}

^a School of Chemistry and Chemical Engineering, Jiangsu University, 301 Xuefu Road, Zhenjiang, 212013, PR China

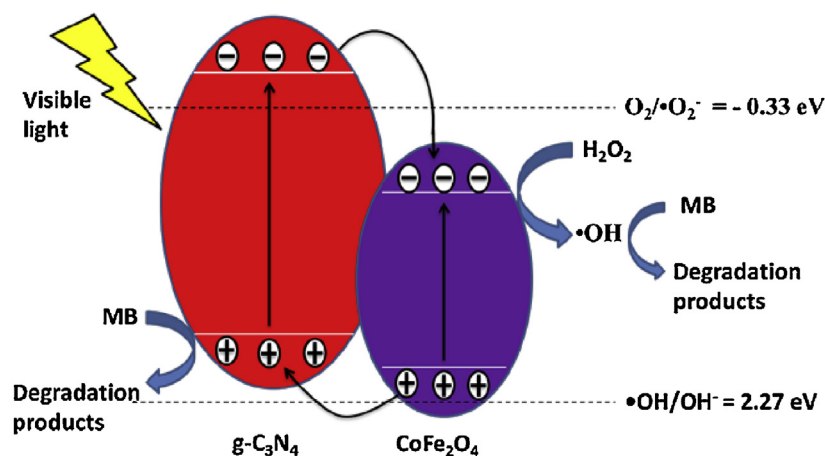
^b School of Pharmacy, Jiangsu University, 301 Xuefu Road, Zhenjiang, 212013, PR China

HIGHLIGHTS

- There was no report to investigate the photocatalytic activities of CoFe₂O₄/g-C₃N₄.
- The CoFe₂O₄/g-C₃N₄ composites are magnetic and stable.
- Combination of CoFe₂O₄ and g-C₃N₄ could enhance the activity of activating H₂O₂.
- The possible photocatalytic mechanism was discussed in detail.

GRAPHICAL ABSTRACT

The CoFe₂O₄/g-C₃N₄ composite could enhance the absorption of visible-light and promote the separation of photogenerated electron-hole pairs. These electrons will activate H₂O₂ to degrade MB.



ARTICLE INFO

Article history:

Received 5 January 2015
 Received in revised form 23 March 2015
 Accepted 25 March 2015
 Available online 2 April 2015

Keywords:

CoFe₂O₄
 g-C₃N₄
 Photocatalyst
 Magnetic
 Visible light

ABSTRACT

A magnetic photocatalytic CoFe₂O₄/g-C₃N₄ composite was successfully synthesized by a simple calcination method. Powder X-ray diffraction (XRD), transmission electron microscopy (TEM), infrared (IR) spectra, UV–Vis diffuse reflection spectroscopy (DRS) and X-ray photoelectron spectroscopy (XPS) were applied to characterize the as-prepared samples. The magnetic properties of CoFe₂O₄ and CoFe₂O₄/g-C₃N₄ composites were measured by using a vibrating sample magnetometer (VSM) at room temperature. Photocatalytic property of the CoFe₂O₄/g-C₃N₄ composite was assessed by degrading methylene blue (MB) in aqueous medium under visible light irradiation. The results showed that the composite of 41.4% CoFe₂O₄/g-C₃N₄ exhibited the highest photocatalytic activity. It could activate H₂O₂ to degrade MB up to 97.3% in 3 h under the visible light irradiation. This enhancement could be attributed to the synergistic effect between CoFe₂O₄ and g-C₃N₄, which could enhance their activity of activating H₂O₂ to degrade MB under visible light. The CoFe₂O₄/g-C₃N₄ composites also have a strong magnetic ability. After the photocatalytic reaction, it can be quickly separated from the water by an extra magnetic field. Moreover, a possible photocatalytic mechanism was proposed.

© 2015 Elsevier B.V. All rights reserved.

* Corresponding author.

** Corresponding author. Tel.: +86 051188791108; fax: +86 051188791108.

E-mail addresses: jbmwgc@126.com (M. He), lhm@ujs.edu.cn (H. Li).

1. Introduction

Photocatalytic technology has been widely used in many fields, including solving the energy problem, decomposing the synthetic dyes in water, eliminating greenhouse effect and so on [1–6]. Nowadays, much attention has been paid to synthesize environment-friendly photocatalysts by the scientific community. With great deal of devotion, tremendous achievements have been scored and lots of semiconductors have been established. TiO_2 is the earliest investigated photocatalyst, it has a series of advantages, such as high chemical resistance, good photostability, environment friendly, easily obtained, inexpensive price and so on [7–10]. However, TiO_2 has one big disadvantage, which is the large band gap of 3.2 eV. It only can be excited by wavelengths below 388 nm. As we all know, the UV region is only up to ~4% of the solar which strongly restrict the application of TiO_2 [11]. Great efforts have been taken out to extend the application of TiO_2 , including chemical modifications, even finding new semiconductors to replace it [12].

Graphitic carbon nitride ($g\text{-C}_3\text{N}_4$), a new semiconductor photocatalyst, has attracted increasing attention and has been applied in many energy-related fields in recent years. Due to its unique electron configuration, special optical structure, highly thermal stability [13,14], it has been frequently used in oxygen reduction reactions [15], heterogeneous catalysis [16], water splitting, photodegradation of organic pollutants [17] and many other fields [18,19]. Besides, $g\text{-C}_3\text{N}_4$ is nontoxic and inexpensive. It could be easily obtained by the thermal condensation of nitrogen-rich precursors, such as cyanamide, dicyandiamide [20]. However, $g\text{-C}_3\text{N}_4$ still has shortcomings, including high recombination rate of the photogenerated electron–hole, low surface area, and low conductivity [21]. Fortunately, the good stability of $g\text{-C}_3\text{N}_4$ makes it easy to be modified by chemical and physical methods. Wang et al. [22] prepared $g\text{-C}_3\text{N}_4$ nanorods by nanoconfinement and promotion of crystallinity on enhancing its photocatalytic conversion ability. It has been reported that the combination of $g\text{-C}_3\text{N}_4$ and other semiconductors could extend the absorption range of $g\text{-C}_3\text{N}_4$. Meanwhile, heterojunctions were formed between different semiconductor materials [23–26], which could greatly improve the separation of electron–hole pairs and gain better photocatalytic activity. Zhu et al. [27] fabricated core/shell structured $\text{C}_3\text{N}_4/\text{BiPO}_4$ photocatalyst via a facile ultrasonic dispersion method and the photocatalyst displayed dramatic photocatalytic activity. Additionally, lots of other methods also have been established to improve the photocatalytic performance and optical absorption of $g\text{-C}_3\text{N}_4$, such as metal deposition and nonmetal doping [28–30]. All these strategies can extend its applications. However, nanostructure $g\text{-C}_3\text{N}_4$ and bulk $g\text{-C}_3\text{N}_4$ both face another problem, which is difficult to be separated after the photocatalytic reaction. An efficient and facile way to separate the catalyst from the suspension is to combine $g\text{-C}_3\text{N}_4$ with a magnetic material. Ye et al. [31] fabricated magnetically separable carbon nitride photocatalyst ($\text{Fe}_3\text{O}_4/g\text{-C}_3\text{N}_4$) and used it for the degradation of Rhodamine B. Although it has met the requirement of separating the photocatalysts from the solution, the direct contact of the two materials leads to the decrease of photocatalytic activities and stability because of the photodissolution of the Fe_3O_4 magnetic particles under the irradiation [32]. There was magnetic $g\text{-C}_3\text{N}_4$ composite has been reported [33], but finding out a stable magnetic material, which could combine with $g\text{-C}_3\text{N}_4$ more facily is still of necessity.

Spinel CoFe_2O_4 has several features, such as high electromagnetic performance, excellent chemical stability, remarkable mechanical hardness and ferromagnetic behaviors [34]. In recent years, it has been reported that CoFe_2O_4 could be used as photocatalysts [35]. Although CoFe_2O_4 is not responsible for photocatalytic reaction [36], it will exhibit very high photocatalytic activity

when combined with conductors. Wang et al. [37] fabricated magnetically separable CoFe_2O_4 -graphene photocatalyst via a facile one-step straightforward hydrothermal strategy and this photocatalyst showed dramatic active catalyst for the degradation of methylene blue (MB), Rhodamine B (RhB), methyl orange (MO). CoFe_2O_4 also has been used to modify other photocatalytic semiconductors and showed synergistic effect, such as $\text{CdS}/\text{CoFe}_2\text{O}_4$ [38], $\text{ZnO}/\text{CoFe}_2\text{O}_4$ [39], $\text{ZnS}/\text{CoFe}_2\text{O}_4$ [40]. Another advantage of CoFe_2O_4 is that it could be used in Advanced Oxidation Processes (AOPs) [41], which has been proven to be an effective way in removing organic pollutants. Costa et al. [42] reported that the introduction of cobalt into magnetite structure to form $\text{Co}_x\text{Fe}_{3-x}\text{O}_4$ could strongly promote the activation of H_2O_2 to degrade the target pollutant. Deng et al. [43] prepared CoFe_2O_4 magnetic nanoparticles and used them for the activation of oxone to generate sulfate radicals for the degradation of diclofenac.

Above all, as a magnetic material, CoFe_2O_4 has emerged a great advantage in the application of degrading organic pollutants. Besides, it has been reported that $g\text{-C}_3\text{N}_4$ could activate H_2O_2 to degrade organic pollutants under visible light irradiation [44]. Therefore, the combination of CoFe_2O_4 and $g\text{-C}_3\text{N}_4$ could be beneficial, since it would fabricate an efficient, stable and magnetic photocatalyst to activate H_2O_2 for removing organic pollutants. Up to now, there is no report about CoFe_2O_4 supported by $g\text{-C}_3\text{N}_4$. Herein, we fabricated magnetic photocatalyst $\text{CoFe}_2\text{O}_4/g\text{-C}_3\text{N}_4$ composites by using a simple calcination method and used it for activating H_2O_2 to decompose MB under visible light irradiation. The $\text{CoFe}_2\text{O}_4/g\text{-C}_3\text{N}_4$ composites have a strong magnetic property, which ensure it can be separated from the water easily. In addition, the combination of CoFe_2O_4 and $g\text{-C}_3\text{N}_4$ can enhance the ability of activating H_2O_2 . At last, a possible photocatalytic mechanism was also proposed.

2. Experiment section

2.1. Synthesis of $g\text{-C}_3\text{N}_4$

The $g\text{-C}_3\text{N}_4$ was prepared with a directly heating dicyandiamide method [45,46].

2.2. Synthesis of CoFe_2O_4

The CoFe_2O_4 was prepared via a sol–gel method [43]. In a typical procedure, 4.0402 g $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ and 1.4553 g $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ were dissolved in 100 mL distilled water (solution A). 2.8821 g citric acid was dissolved in 100 mL distilled water (solution B) and the metal ($\text{Co}^{2+} + \text{Fe}^{3+}$)/citric acid molar ratio was 1. Then solution A was dropwise added into solution B under vigorous magnetic stirring. The mixed solution reacted for 1 h at 60 °C and then dried at 90 °C, the resulting powder was then put into an alumina crucible and calcined for 4 h at a temperature of 400 °C.

2.3. The fabrication of $\text{CoFe}_2\text{O}_4/g\text{-C}_3\text{N}_4$ composites

Samples with different mass fraction were synthesized by a facile calcination method. A typical experiment for the synthesis of $\text{CoFe}_2\text{O}_4/g\text{-C}_3\text{N}_4$ was as follows: 0.1 g CoFe_2O_4 and different amount of $g\text{-C}_3\text{N}_4$ were mixed homogeneous by grounding in an agate mortar at least 20 min, then the mixed powder was put into an alumina crucible and calcined for 4 h at a temperature of 400 °C. According to the TG analysis results, the mass contents of CoFe_2O_4 in $\text{CoFe}_2\text{O}_4/g\text{-C}_3\text{N}_4$ were estimated to be 25.3%, 41.4% and 64.7%, respectively. The different mass fraction of CoFe_2O_4 in the $\text{CoFe}_2\text{O}_4/g\text{-C}_3\text{N}_4$ was named as 25.3% $\text{CoFe}_2\text{O}_4/g\text{-C}_3\text{N}_4$, 41.4% $\text{CoFe}_2\text{O}_4/g\text{-C}_3\text{N}_4$ and 64.7% $\text{CoFe}_2\text{O}_4/g\text{-C}_3\text{N}_4$.

Download English Version:

<https://daneshyari.com/en/article/592219>

Download Persian Version:

<https://daneshyari.com/article/592219>

[Daneshyari.com](https://daneshyari.com)