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Highly active magnetic bismuth tungstate/magnetite composite under visible light irradiation in the presence of hydrogen peroxide

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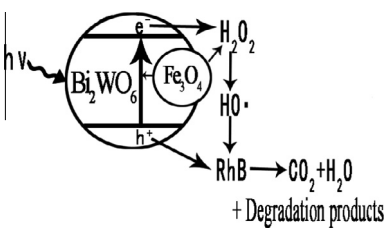
GRAPHICAL ABSTRACT



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ABSTRACT

Bi_2WO_6/Fe_3O_4 composites were synthesized using hydrothermal method and their photocatalytic activity to degrade rhodamine B (RhB) under visible light irradiation assisted with H_2O_2 and underlying mechanisms were investigated. The composites were 3D flower-like microspheres constructed by 2D Bi_2WO_6 nanosheets loaded with spherical Fe_3O_4 nanoparticles. The composited photocatalysts could be easily harvested from the reaction solution by an external magnetic field. In the presence of H_2O_2, a large amount of hydroxyl radicals (·OHs) were produced by H_2O_2 reacting with photogenerated electrons. Fe_3O_4 not only promoted the separation of hole–electron pairs but also acted as a Fenton-like reagent, expediting the production of ·OH. Thus, the composites in the presence of H_2O_2 displayed much higher photocatalytic efficiency to degrade RhB than pure Bi_2WO_6. 98% of RhB (initial concentration 10 mg/L) was degraded in 2 h visible irradiation and 60% of total organic carbon (TOC) was removed in 3 h by 0.5 g/L Bi_2WO_6/Fe_3O_4 (5:1). The composite displayed high photodegradation efficiency at pH 3–9. Our study suggests that a visible light driven and highly active magnetic photocatalyst-Fenton coupling oxidation system may have potential application in water treatment and environmental cleaning.

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1. Introduction

In recent years, semiconductor photocatalytic process has found its wide application in water treatment as a low-cost, environmental friendly and sustainable treatment technology [1]. TiO_2 is one of the most widely applied photocatalysts under irradiation of ultraviolet light. However, TiO_2 can only absorb ultraviolet wavelength, which constitutes only ~3–5% of solar light [2]. In order to make better use of sunlight in nature, efforts have been done to develop catalysts which are active to visible light. Bismuth photocatalysts, including Bi_2WO_6, are active to visible light and display potential photocatalytic efficiency to many contaminants and azo dyes under visible light irradiation [3–7]. As a visible-light-driven photocatalyst, Bi_2WO_6 displays potential catalytic capacity to many organic chemicals under irradiation of visible light and it is primarily synthesized by hydrothermal or solvothermal method [8]. In order to improve catalytic efficiency, Bi_2WO_6 catalysts with

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different morphology and crystal structures were synthesized [8,9]. Another effective technique is to dope Bi_2WO_6 with metal, or metal oxide, which might trap the photogenerated electrons and restrain the recombination of hole–electron pair [10–13].

However, one of the shortcomings which limit the real application of these catalysts in water treatment is that it is very difficult to separate the catalysts from the reaction solution at the end of reaction without the assistance of centrifugation or filtration. Recently, many studies have been conducted to prepare photocatalysts with magnetic materials, which could be separated and recycled conveniently by applying an external magnetic field. Many magnetic iron materials, such as $CoFe_2O_4$ [14], $NiFe_2O_4$ [15], Fe_3O_4 [16], were coupled with different photocatalysts to prepare composites with magnetic property. In many cases, doping with magnetic materials would reduce photocatalytic activities of the original catalysts due to decreasing activity sites available for reaction. Strategies are needed to prepare magnetic composites which retain the good photoactivity of the original catalyst and obtain good magnetic property.

Magnetite (Fe_3O_4) is an ideal magnetic material and widely used in catalysis because of its low cost and easy preparation [17]. Besides its ultrahigh-density magnetic property, Fe_3O_4 has been used as heterogeneous Fenton-like catalyst for catalytic oxidation of organic compounds [18,19]. H_2O_2 is a typical oxidative agent and has been widely used in practical water treatment [20]. As an electron capture agent, H_2O_2 can react with photogenerated electrons to produce hydroxyl radicals (·OHs) [4,21].

$$H_2O_2 + e^-_{CB} \rightarrow {}^{\cdot}OH + OH^- \quad (1)$$

Thus, when H_2O_2 is co-present with Bi_2WO_6/Fe_3O_4 composite, it might interact with the composite and affect the photocatalytic capacity. It was reported that Bi_2WO_6 and its composites such as Co_3O_4/Bi_2WO_6, C/Fe–Bi_2WO_6, Bi_2WO_6@carbon/Fe_3O_4 exhibited higher photodegradation efficiency with the assistance of H_2O_2 [21–24]. Xu et al. and Liu et al. synthesized Bi_2WO_6/Fe_3O_4 composites and achieved high degradation efficiency for RhB [25,26]. However, the mechanisms involved in the complex system were not fully investigated.

The current study aimed to dope Bi_2WO_6 with Fe_3O_4 to prepare magnetic Bi_2WO_6/Fe_3O_4 composites and investigate the combined degradation mechanisms of Bi_2WO_6/Fe_3O_4 in the presence of H_2O_2. Rhodamine B (RhB) was used to imitate nonbiodegradable, toxic organic compounds with multiple benzene rings. The photocatalytic activity of the magnetic composites to RhB under visible light ($\lambda > 400$ nm) in the presence of H_2O_2 was evaluated. The respective roles of Bi_2WO_6, Fe_3O_4 and H_2O_2 and the reaction mechanism were investigated extensively. The impacts of H_2O_2 concentration and solution pH on the photocatalytic performance were studied. The degradation pathway was also investigated. The proposed strategy was useful for designing a visible light driven and magnetic photocatalyst-Fenton coupling oxidation system for wastewater treatment.

2. Materials and experiment

2.1. Materials and reagents

Rhodamine B (RhB) (98%) was purchased from J & K Scientific Ltd. (Beijing, China). $FeCl_3{\cdot}6H_2O$ (analytical reagent, AR) was bought from Guangfu Technology Co. Ltd. (Tianjiin, China). Sodium acetate (AR), NaOH (AR) and HNO_3 (65%) were purchased from Jiangtian Chemical Technology Co. Ltd. (Tianjin, China). $Bi(NO_3)_3{\cdot}5H_2O$, $Na_2WO_4{\cdot}4H_2O$ (AR) and KI (Guaranteed reagent) was purchased from Yingda Chemical Technology Co. Ltd. (Tianjin, China). Isopropanol (AR) was provided by Fengchuan Chemical Technology Co. Ltd. (Tianjin, China). $AgNO_3$ (AR) was obtained from Tairuier Chemical Co. Ltd. (Shanghai, China). Glycol and absolute ethyl alcohol (AR) were purchased from Concord Technology Co. Ltd. (Tianjin, China). Terephthalic acid (TPA) (99%) was purchased from ACROS (New Jersey, USA).

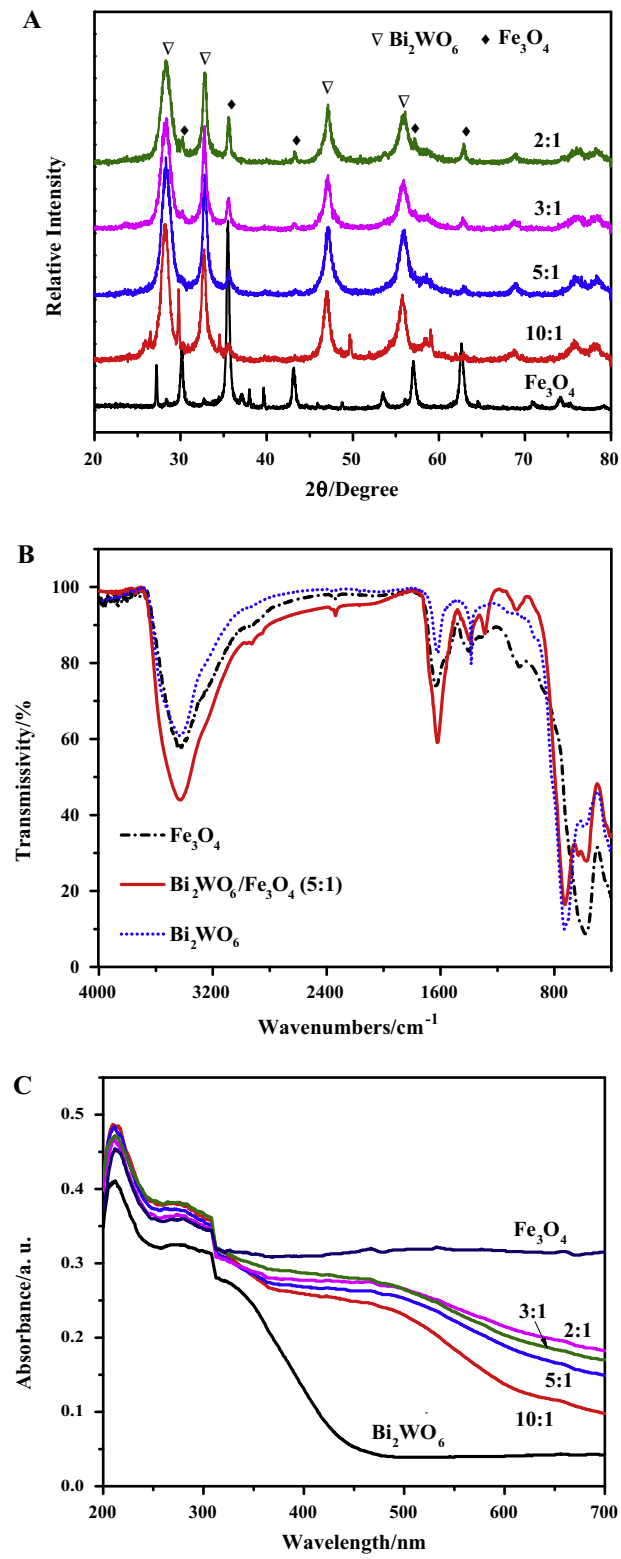

Fig. 1. (A) The XRD patterns of Fe_3O_4 and the prepared Bi_2WO_6/Fe_3O_4 composites (2:1, 3:1, 5:1, 10:1); (B) The FT-IR spectra of Fe_3O_4, Bi_2WO_6/Fe_3O_4 (5:1) and pure Bi_2WO_6; (C) UV–vis diffuse reflectance spectra of the prepared Bi_2WO_6, Fe_3O_4 and Bi_2WO_6/Fe_3O_4 composites (2:1, 3:1, 5:1, 10:1).

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