



Synergetic effect of carbon sphere derived from yeast with magnetism and cobalt oxide nanochains towards improving photodegradation activity for various pollutants

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ABSTRACT

Developing high-efficiency and low-cost semiconductor photocatalysts with carbon layer modification has been proved to possess the potential advantage for dramatically improved photocatalytic performance in recent years. Here, a novel magnetic core-shell Fe₃O₄@C modified with Co₃O₄ nanochains photocatalyst (Fe₃O₄@C/Co₃O₄) is first constructed via a facile synthetic strategy using inexpensive and ordinary yeast as carbon source. The obtained composite structure is conducive to the transfer and separation of charge carriers due to the synergetic effect of carbon layer and Co₃O₄ nanochains. In consequence, the photodegradation activity of various pollutants is significantly enhanced relative to that of pure Co₃O₄ under visible light irradiation. Moreover, the introduction of magnetic materials makes this photocatalyst recycled easily by an external magnetic field and reused effectively without obvious activity loss. This work may provide a feasibility analysis and exemplificative strategy for using simple raw materials as carbon source to observably improve universal photocatalytic activity of composite semiconductor photocatalyst.

1. Introduction

Over the past few years, water pollution is one of the most serious environmental problems, which becomes a formidable challenge to the sustainable development of modern human society. Semiconductor photocatalysis, as a kind of green technology, has achieved rapid development attributing to its potential advantages for solving the environmental pollution problem and energy crises [1–3]. As far as semiconductor photocatalysts are concerned, the photocatalytic activity of single semiconductor isn't very high generally. Thus, various modification strategies have been developed to further improve the photocatalytic performance, including doped with foreign elements [4–6], surface decoration with noble metal [7–9], constructed heterojunction with other energy-match semiconductor [10–13] and so on. Among those modified methods, carbon modified have attracted great attentions in recent years because of its unique superiorities contributing to the improvement of photocatalytic activity, such as the superior electrical conductivity, exceptional thermal properties, good light

absorption performance and so on. For example, Li et al. prepared carbon modified BiOX photocatalyst by one-step hydrolysis method using biochar as carbon source [14]. Core-shell structure C@CdS with a thin coating layer of amorphous carbon has also been prepared by one-pot hydrothermal carbonization method in the presence of glucose [15,16]. The improved photocatalytic activity of them confirmed that the introduced carbon material was an available modified way for semiconductor photocatalyst. Nevertheless, the carbon source they used always involves glucose, dimethylglyoxime, organic acid, maize cobs, etc., which may increase cost and complex preparation process as well as even cause secondary environment pollution. Therefore, it is necessary for exploiting a cost-effective, simple and eco-friendly carbon source to improve photocatalytic activity and other physicochemical property. Microorganism carbon may have a great foreground for this application due to its own advantages such as low cost, reproducible and abundant. It has been reported that the yeast carbon modified TiO₂ not only has the biosorption ability of yeast but also improves the photocatalytic activity of TiO₂ obviously [17–19]. Song et al. [20] also

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obtained TiO₂-yeast carbon hybrid microspheres with controllable structures as photocatalyst. Those results demonstrated yeast can be an ideal choice as carbon source for photocatalyst modification. However, TiO₂ can be only excited by the ultraviolet light due to its wide band gap (3.2 eV) and thus limits its practical application [21,22]. Therefore, designing and constructing of novel carbon modified visible-light semiconductor using yeast as carbon source is desired and significant to achieve high-performance photocatalysts.

As is well known, transition metal oxides have extensive application in the photocatalytic field due to the variety advantages of high stabilization, friendly environment, low cost and even visible-light photocatalytic activity [23,24]. Especially, cobalt oxide (Co₃O₄) is a good candidate in photocatalytic field because of its interesting electronic structure, good light absorption performance and high thermal and chemical stability. However, pure Co₃O₄ has a relatively low photocatalytic activity due to lower carrier separation efficiency [25]. Considering the above advantages of yeast as carbon source, we suspect if yeast was introduced into Co₃O₄ system, it may be an effective way to improve its photocatalytic activity dramatically for practical application. Referring to the practical application of photocatalyst, it is very necessary to effectively separate and recycle the used photocatalyst to avoid secondary pollution and keep stability [26–28]. It is reported that core-shell structures with a magnetic core endue the materials with magnetic property and show superior separation effect [29,30]. Therefore, the incorporation of carbon and magnetic materials into the Co₃O₄ system may be a very effective to greatly enhance photocatalytic activity and separate characteristics of Co₃O₄, which can further promote the value of practical application.

Herein, a novel carbon modified Co₃O₄ nanochains photocatalyst (Fe₃O₄@C/Co₃O₄) by using yeast as carbon source is successfully prepared by a facile and eco-friendly synthetic strategy for the first time. In comparison with the pure Co₃O₄, the Fe₃O₄@C/Co₃O₄ exhibits superior universal photocatalytic degradation activity for various contaminants in the water under visible light, including antibiotic, heavy metal Cr(VI) and colored dye RhB. It derived from the synergetic effect of each function part in photocatalyst, in which the electrical conductivity of folium carbon facilitates electrons transfer from cobalt oxide. Additionally, the as-prepared photocatalysts could be easily separated from reaction solution by an external magnetic field. Therefore, this work extends the development of carbon source as carbon layer modification with other visible-light semiconductor photocatalyst to further improve universal photocatalytic activity and reusability for practical applications.

2. Experimental

2.1. Materials

The powdered yeast was purchased from Angel Yeast Company. Other reagents including ferric chloride hexahydrate (FeCl₃·6H₂O), ethylene glycol (EG), diethylene glycol (DEG), sodium acrylate (CH₂=CHCOONa, Na acrylate), sodium acetate (CH₃COONa, NaOAc), citric acid, cetyltrimethylammonium bromide (CTAB), 1-hexanol, hexane, cobalt acetate, ammonium oxalate, chloroform, methanol, benzoquinone (BQ), isopropanol (IPA), triethanolamine (TEOA), 5,5-dimethyl-1-pyrroline N-oxide (DMPO) were obtained from Aladdin. Ethanol (C₂H₅OH, 95.0%) was purchased from Sinopharm Chemical Reagent Co.,Ltd. Tetracycline (TC), Cr (IV) and RhB dye were analytical pure and used without further purification, distilled water was used in the whole experiments.

2.2. Synthesis

2.2.1. Synthesis of oxalate precursor

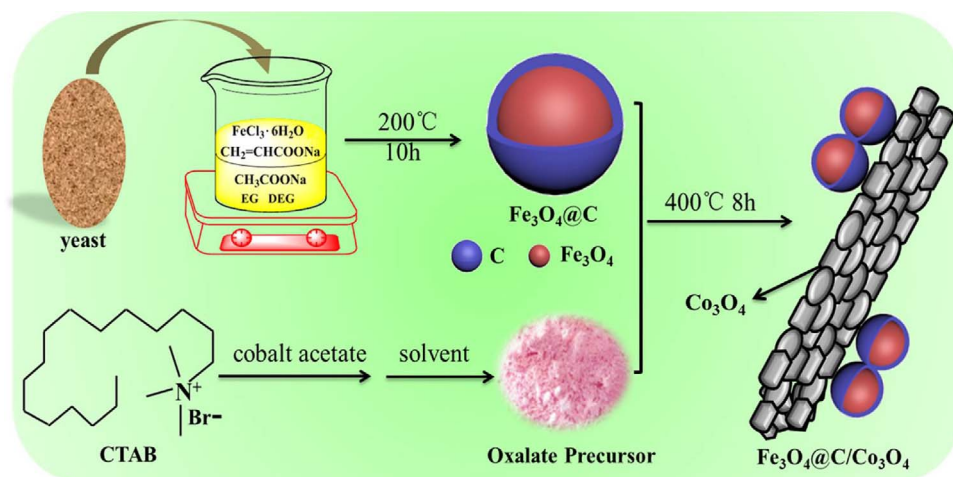
The synthesis of oxalate precursor is according to the reference [31]. Typically, microemulsions containing cetyltrimethylammonium bromide (CTAB, 2.0 g) as a surfactant, 1-hexanol (20 mL) as cosurfactant and hexane (35 mL) as the lipophilic phase were first prepared and were mixed separately with an aqueous solution of 0.1 M cobalt acetate and 0.1 M ammonium oxalate. Both microemulsions were mixed slowly and stirred overnight. The pink precipitate then obtained was centrifuged and washed with a 1/1 mixture of chloroform and methanol (200 mL) and subsequently dried at 60 °C for 12 h.

2.2.2. Synthesis of magnetic core-shell Fe₃O₄@C

Yeast (0.1 ~ 0.5 g), FeCl₃·6H₂O (2.4 g), CH₂=CHCOONa (3.4 g), NaOAc (3.4 g) were added into a mixture of EG (22.5 mL) and DEG (22.5 mL) under ultrasonication for about 1 h. The homogeneous solution obtained was transferred to a Teflon-lined stainless-steel autoclave and sealed to heat at 200 °C. After reaction for 10 h, the autoclave was cooled to room temperature. The Fe₃O₄@C obtained was washed several times with ethanol and water, and then dried in vacuum at 65 °C for 10 h.

2.2.3. Synthesis of magnetic core-shell Fe₃O₄@C/Co₃O₄ nanochains

Fe₃O₄@C and cobalt oxalate precursor were added into EG under magnetic stirring for about 1 h. Then the samples with deionized water and ethanol washing several times and dried in vacuum at 65 °C for 10 h. Finally, the samples were heated to 400 °C at a rate of 2 °C/min in dry synthetic air (20% O₂, 80% N₂) and kept at 400 °C for 8 h in a tubular furnace and then cooled to ambient temperature normally. The



Scheme 1. Schematic illustration of the preparation process of the Fe₃O₄@C/Co₃O₄ sample.

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