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Controlled fabrication of bismuth vanadium oxide hierarchical microtubes with enhanced visible light photocatalytic activity

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ABSTRACT

Monoclinic bismuth vanadium oxide ($BiVO_4$) hierarchical microtubes self-assembled by several nanowires have been successfully fabricated by a template-free approach. The morphology and structure of the as-prepared sample were characterized by scanning electron microscopy (SEM), X-ray powder diffraction (XRD) and UV–vis spectroscopy. $BiVO_4$ microtubes have monoclinic structure with energy band gap of 2.36 eV. Based on the time-dependent experiments, the possible formation mechanism of $BiVO_4$ microtubes was proposed. In addition, the photocatalytic activity of the as-prepared $BiVO_4$ samples was evaluated by the degradation of methyl orange (MO) and various reactive dyes (RDs) under xenon lamp irradiation. In comparison with $BiVO_4$ microrods, $BiVO_4$ microtubes with hollow and porous structures showed higher photocatalytic activities and universality under visible-light irradiation. Due to the unique hierarchical hollow structures contributing to the increase of photocatalytic activities, $BiVO_4$ microtubes exhibited the potential application as the promising visible-light photocatalyst.

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1. Introduction

Hollow semiconductors with micro- and nano-structures have attracted considerable attention for its applications in the fields of rechargeable batteries [1], sensors [2], catalysis [3], and target drug delivery [4]. In comparison with solid materials, hollow semiconductors have exhibited unique characteristics including low density, high surface area, distinct optical properties, low coefficients of thermal expansion and refractive indices [5,6]. In the recent years, dyes and pigments from various industrial branches have caused severe environmental contamination due to the emission of the toxic and colored wasterwater into water bodies. The presence of low concentrations of dyes in effluent streams seriously affected the nature of water, inspiring fundamental and applied research interest in the area of environmental remediation [7,8].

Using semiconductors as photocatalysts can be an alternative to conventional methods for the removal of organic pollutants from water [9,10]. The photocatalytic activities of semiconductors with tailored structures are getting more and more attention due to the unique electronic structure composed of a filled valence band and an empty conduction band. Among the semiconductors employed, titanium dioxide (TiO_2) is the most extensively used photocatalyst due to its high photocatalytic activity, non-toxic nature, excellent chemical and mechanical stability [11,12]. However, TiO_2 can only utilize a small ultraviolet (3%–4%) irradiation of the solar spectrum due to its wide band gap of 3.2 eV. In order to increase the utilization of solar energy, exploring visible-light sensitive catalysts is the most urgent issue for the removal of organic pollutants.

Bismuth vanadium oxide ($BiVO_4$), as a ternary oxide semiconductor, has been recognized as an effective visible-light-driven photocatalyst for the degradation of organic pollutants in wastewater due to the interaction between 6s Bi and 2p O orbitals at the top of the valence band [13–20]. $BiVO_4$ have three crystalline phases, namely, tetragonal scheelite, tetragonal

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zircon and monoclinic scheelite. However, among the possible structures, only the monoclinic $BiVO_4$ with the band gap energy of 2.4 eV has good photocatalytic ability under visible-light irradiation [13,20]. Not only the structures, the photocatalytic properties also strongly depend on the morphology of the crystal. Therefore, $BiVO_4$ crystals with various micro- and nanostructures, such as nanosheets [13], hollow spheres [14], fibers [15], mesopore [16], starlike structures [17], microtubes [18–20], etc. have been fabricated and studied by many research groups all over the world to enhance the photocatalytic activity. For example, monoclinic $BiVO_4$ nanosheets by hydrothermal method assisted by sodium dodecyl benzene sulfonate (SDBS) as a morphology directing template showed good visible photocatalytic activities under solar irradiation [13]. The novel core–shell structured $BiVO_4$ hollow spheres with an ultrahigh specific surface area via a one-pot, surfactant- and template-free hydrothermal route exhibited a superior photocatalytic activity over various of morphological products in the photodegradation of rhodamine B under visible-light irradiation [14]. Nanoplate-stacked star-like monoclinic $BiVO_4$ fabricated by a hydrothermal method [17] and single-crystalline $BiVO_4$ microtubes with flower-like morphology by a facile reflux method [18] also exhibited the higher visible-light-driven photocatalytic efficiency than the reference $BiVO_4$ sample prepared by solid-state reaction.

As is well known, the catalytic activity can be effectively enhanced by reducing the particle size, while this would cause the agglomeration of nanoparticles with small size and then lead to the decrease in activity. Therefore preventing the agglomeration of nanoparticles is extremely important to develop $BiVO_4$ photocatalytic performance. In order to resolve the above problem, designing and assembling $BiVO_4$ hierarchical hollow structures by nanomaterials may be an effect approach for the improvement of photocatalytic activity. This is due to the fact that the hierarchical hollow materials not only have the characterizations of nanomaterials with high activity, but also have the features of hollow structures with large internal cavity, helpful for the entry of reactants and the release of products. At the same time, the controlled design of hierarchical structures can effectively prevent the agglomeration of nanoparticles. Therefore, it is of great significance to explore the preparation of the novel $BiVO_4$ hierarchical hollow structures constructed by nanobuilding blocks with the view of the improvement of visible light photocatalytic activity.

Template-free synthesis could be helpful to make the products with high crystallization, narrow grain size-distribution and high purity in the absence of heat treatment at high temperature. Therefore, it is usually considered as an idea technique of synthesizing promising $BiVO_4$ materials with special morphology. In this paper, we describe a facile template-free approach to fabricating monoclinic $BiVO_4$ hierarchical microtubes self-assembled by nanowires. Although compared with other synthesis method of $BiVO_4$ microtubes [18–20], our method is possibly not the best, but it provides another approach for the fabrication of monoclinic $BiVO_4$ microtubes by using the template-free approach. At the same time, it is very facile and effective for preparing the tubular materials with high yield. In order to make clear the effect of hollow structure on the photocatalytic activity, the solid $BiVO_4$ microrods have been effectively fabricated as comparison. And the as-prepared $BiVO_4$ microtubes exhibited higher photocatalytic efficiency and universality in the degradation of methyl orange (MO) and various reactive dyes (RDs) under visible-light irradiation.

2. Experimental details

2.1. Reagents

Bismuth nitrate ($Bi(NO_3)_3 \cdot 5H_2O$), ammonium metavanadate (NH_4VO_3), sodium hydrogen carbonate ($NaHCO_3$), cetyltrimethyl ammonium bromide (CTAB), hydrogen peroxide, and all other regents are analytical grade and used without further purification. Methyl orange (MO) and reactive dyes (RDs), such as reactive dark blue R-2GLN (RDB), reactive red R-4BD (RR), reactive black R-2BR (RB) and reactive brilliant yellow R-4GLN (RBY), were purchased from Zhejiang Runtu Co., Ltd., China. Deionized water was used throughout the experiments.

2.2. Synthesis of $BiVO_4$ samples

A typical synthesis of $BiVO_4$ microtubes was shown in the following way: 1 mmol of $Bi(NO_3)_3 \cdot 5H_2O$ and 1 mmol of NH_4VO_3 were dissolved in 20 mL nitric acid (1.0 mol L^{-1}) under stirring. After dissolution, 0.5 g of CTAB and 1.5 g of $NaHCO_3$ were added into the above solution in turn. The mixed solution was stirred for another 0.5 h and then sealed in a Teflon-lined stainless-steel autoclave (25 mL capacity). The autoclave was heated to and maintained at 80 °C for 8 h, and then allowed to cool to room temperature. The light yellow products were collected by centrifugation and rinsed more than five times with deionized water and ethanol, and dried in air at 60 °C for 12 h. The solid $BiVO_4$ microrods were prepared by the similar method but at the reaction temperature of 120 °C for 8 h without the addition of CTAB.

2.3. Characterization

The morphology of the synthesized $BiVO_4$ samples was characterized by scanning electron microscopy (SEM, JEOL JSM-6360 LV SEM). X-ray powder diffraction (XRD) pattern was obtained on a Empyrean X-ray diffract meter using Cu $K\alpha$ radiation source (λ=0.15406 nm) to determine the crystal phase of the obtained sample. Nitrogen sorption isotherms and pore size distribution of $BiVO_4$ microtubes were obtained from Micromeritics TriStar 3020 instrument. UV–vis diffused reflectance spectra of the samples were measured by a UV–vis spectrophotometer (UV-2550, Shimadzu, Japan). $BaSO_4$ was used as a reflectance standard.

2.4. Photocatalytic activity

Visible light photocatalytic activities of the obtained photocatalysts were evaluated by the decomposition of MO and RD solution (reactive dark blue R-2GLN, reactive red R-4BD, reactive black R-2BR and reactive brilliant yellow R-4GLN) in an aqueous solution at ambient temperature. A 250-W xenon arc lamp was used as the irradiation source. Photoreaction was performed in a quartz tube containing photocatalyst (10 mg) and 20 mg L^{-1} MO or RD solution

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