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Short communication

Room temperature aqueous synthesis of $BiVO_4/NaBiO_3$ heterojunction with high efficiency for sulfadiazine removal

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ABSTRACT

A novel $BiVO_4/NaBiO_3$ composite catalyst was firstly prepared in room-temperature aqueous, which was a facile and controlled synthesis approach. The as-prepared $BiVO_4/NaBiO_3$ composite samples exhibited excellent photocatalytic activity and recyclability for sulfadiazine degradation under xenon light irradiation. The photocatalytic mechanism was discussed based on the reactive species analysis.

1. Introduction

Sulfadiazin (SDZ) belongs to one kind of sulfonamide antibiotics, which is extensively used in both human and veterinary medicine. A large number of antibiotics and metabolites are being continuously released into the environment because of increasing antibiotic drugs consumption, which has being gradually become one type of important emerging pollutant and an overwhelming ecological security problem all over the world [1–4]. Many kinds of antibiotics have been increasingly detected in the aquatic environment because the waters are important environment for migration and distribution of pollutants. Meanwhile, people are paying more and more attentions to the associated environmental hazards and modification. As emerging micropollutants, antibiotics are high toxicity but very low concentration in environment. The conventional techniques of physical, chemical and biological methods are difficult to remove antibiotics from waters [5–8]. Therefore, it is very urgent to develop effective techniques to eliminate these emerging pollutants. In recent years, photocatalysis technologies are gradually researched and developed, and semiconductor photocatalysis is considered as a green and promising technology to eliminate pollution under sunlight irradiation. It showed that the light response efficiency is a key factor for the development of semiconductor photocatalytic technology. However, the light response efficiencies of existing photocatalysts are very low. On the one hand, most of the photocatalysts only respond to ultraviolet light; on the other hand, the poor quantum efficiencies and low photocatalytic performances of many photocatalysts are often caused for the rapid recombine of the photogenerated electrons and holes [9–11]. Furthermore, UV light only accounts about 4% and visible light accounts more than 43% of the solar radiation. Therefore, developing of novel photocatalysts enhancing utilization of solar light and separation photoinduced electron–hole pairs are more significant, and build-up of heterojunction photocatalyst is an important strategy [12–14].

$BiVO_4$ is non-titania based photocatalyst, which is attracted extensive research due to its narrow band gap (2.4 eV) and effective photocatalytic activities for water splitting and pollutant removal under visible light or sun light irradiation. Furthermore, $BiVO_4$ is stability and cheap, and the increasing attentions are paying on $BiVO_4$ photocatalysts [15–18]. However, the photocatalytic efficiency of single $BiVO_4$ photocatalyst is also very low. The fast recombination of photoinduced charge carriers is one major limitation of $BiVO_4$ photocatalysis system. To overcome this obstacle, various $BiVO_4$ based photocatalyst composites have been extensively studied, such as $BiVO_4/ZnO$, $BiVO_4/Bi_2WO_6$, $BiVO_4/Bi_2S_3$, $BiVO_4/CaFe_2O_4$ [19–22] and so on. As far as we know, there is no report about $BiVO_4/NaBiO_3$ heterojunction photocatalyst. $NaBiO_3$ is regarded as an alternative attractive visible light response type photocatalyst, which has a band gap of 2.6 eV. It showed that the photocatalytic efficiency of $NaBiO_3$ is still required to be further improved. It was reported that $NaBiO_3$ has more negative E_{CB} and E_{VB} than $BiVO_4$. The energy conduction band (E_{CB}) of $BiVO_4$ and $NaBiO_3$ are 0.31 eV and −0.21 eV, respectively. The valence band (E_{VB}) of $BiVO_4$ and $NaBiO_3$ are 2.78 eV and 2.15 eV, respectively [23–26]. Thus it can be seen that the staggered level heterojunction could be constructed by coupling with $BiVO_4$ and $NaBiO_3$, and the photocatalytic activity could also be enhanced. In the present work, a novel $BiVO_4$/

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$NaBiO_3$ composite was successfully synthesized in room-temperature aqueous, which showed superior photocatalytic efficiency under a xenon lamp irradiation.

2. Experimental

A detailed description of the synthesis, characterization, photocatalytic test, and product analysis are presented in supporting information.

3. Results and discussion

3.1. Characterizations

3.1.1. Crystal structures

The crystalline structure and composition of the synthesized samples were analyzed by X-ray diffraction (XRD). A scanning rate of 1 s/step was applied to record the pattern in the 2θ range of 10–90°. The detection result of XRD patterns of as-prepared samples are presented in Figure SI1 (in the Supporting Information). The prepared $BiVO_4$ was indexed as crystallized monoclinic $BiVO_4$ (JCPDS card no. 14–0688) [23,27]. The 2θ diffraction peaks of 28.8°, 30.5°, and 42.5° can be respectively indexed as (112), (004), and (015) planes of monoclinic $BiVO_4$ structure. The XRD diffraction peaks of the $NaBiO_3 \cdot 2H_2O$ can match with the hexagonal structure of $NaBiO_3 \cdot 2H_2O$ (JCPDS card no. 30–1161) [23,27]. The 2θ diffraction peaks of 11.9°, 18.3°, and 31.9° are three strongest diffraction peaks of hexagonal structure of $NaBiO_3 \cdot 2H_2O$, which correspond to the crystal indexes of (001), (100) and (110), respectively. To confirm the chemical element composition and valence state in the $BiVO_4$/$NaBiO_3$ sample (1:8), the X-ray photoelectron spectroscopy (XPS) was analyzed. The analysis results are showing in Figure SI1. It shows that the peak locates 529.5 eV which corresponds to Na 1 s. The peaks at 158.2 eV and 163.6 eV represent the binding energies of Bi $4f_{7/2}$ and Bi $4f_{5/2}$, respectively. All the Bi species on the surface of $BiVO_4$/$NaBiO_3$ are mainly attributed to Bi^{5+}. The O1s peak of 529.5 eV corresponds to the lattice oxygen (Bi-O) enveloping of $BiVO_4$ and $NaBiO_3 \cdot 2H_2O$. The two peaks at binding energies of 517.0 eV and 524.2 eV are the signals of V2p3/2 and V2p1/2, which correspond to the V^{5+} [25,28]. Thus it can be seen that the $BiVO_4$/$NaBiO_3$ heterojunction composites were successfully synthesized by facile aqueous method at room temperature.

3.1.2. Morphology characteristics

As shown in Fig. 1, the commercial $NaBiO_3$ particles comprise typical flake hierarchical microspheres, which are consisted by a large number of nanosheets with width (length) of about 10 μm. The as-prepared $BiVO_4$ consists of irregular nanoparticles with an average diameter of less than 1 μm. The SEM photographs of $BiVO_4$/$NaBiO_3$ revealed that the apertures of $NaBiO_3$ were filled with many $BiVO_4$ nanoparticles. To further survey the microscopic structure information of the $BiVO_4$/$NaBiO_3$ heterojunction composite (1:8), transmission electron microscopy (TEM) detection was carried out. As displayed in Fig. 1 (TEM $BiVO_4$/$NaBiO_3$), a large amount of $BiVO_4$ nanoparticles were observed in the apertures of $NaBiO_3$ particles. Meanwhile, the specific surface areas of the sample were detected by Brunauer-Emmett-Teller (BET) theory, which revealed the BET results of $BiVO_4$, $NaBiO_3$ and $BiVO_4$/$NaBiO_3$ (1:8) are 20 $m^2\ g^{-1}$, 30 $m^2\ g^{-1}$ and 41 $m^2\ g^{-1}$, respectively.

3.1.3. Optical properties

The UV–vis diffuse-reflectance spectra (DRS) of the as-prepared samples were demonstrated in Figure SI2 (in the Supporting Information), which shows that the $BiVO_4$/$NaBiO_3$ heterojunction composite (1:8) exhibits a stronger absorption in the UV–visible light region. According to the energy band theory of semiconductor, the optical absorption band edge can be evaluated by Kubelka–Munk equation. The band gap of as-prepared samples are calculated and shown in Figure SI2a (the insert). The band gap of $BiVO_4$/$NaBiO_3$ heterojunction composite (1:8) is 2.43 eV, which is between the band gap of $BiVO_4$ (2.39 eV) and that of $NaBiO_3$ (2.51 eV). It suggests that $BiVO_4$/$NaBiO_3$ composite could have higher visible light response ability than the pure $BiVO_4$ and single $NaBiO_3$. The superior sunlight response of $BiVO_4$/$NaBiO_3$ heterojunction

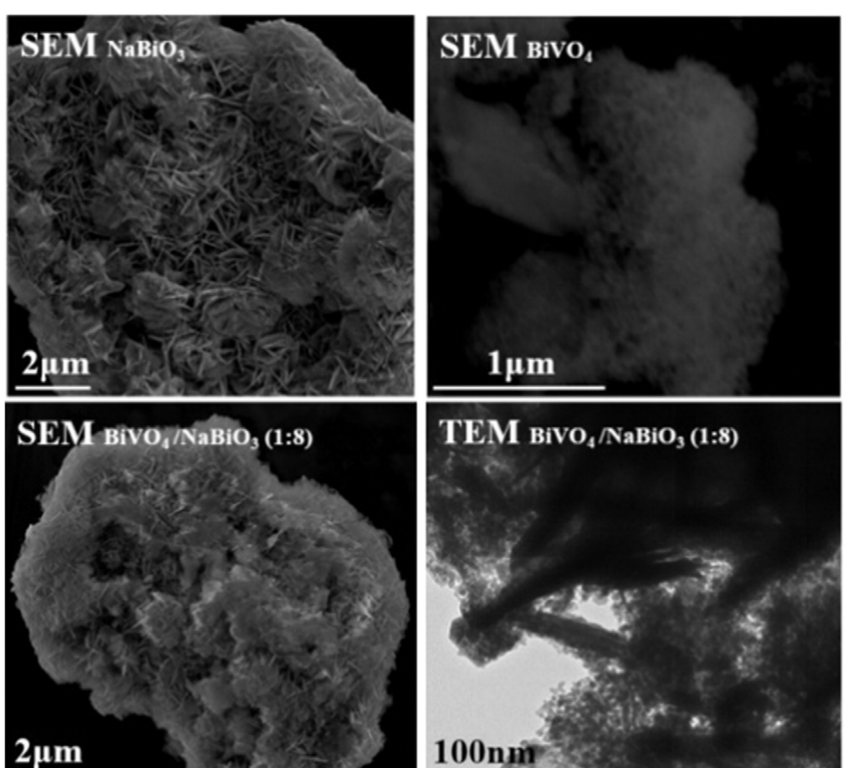

Fig. 1. SEM and TEM figures of the samples.