



NaBiO₃/BiO_{2-x} composite photocatalysts with post-illumination “memory” activity

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ABSTRACT

NaBiO₃/BiO_{2-x} composite photocatalyst was fabricated via a hydrothermal process. As-obtained materials not only exhibited excellent photocatalytic activity toward the degradation of methyl orange (MO) and phenol under visible light irradiation, but also showed strong post-illumination “memory” activity, which enables it to remove the organic pollutants in the dark, promoting the practical application of photocatalyst.

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1. Introduction

Over the past a few decades, photocatalysis has attracted considerable attentions for its environmental applications. Many photocatalysts such as TiO₂, Ag₃PO₄, g-C₃N₄ and Bi₂WO₆ have been developed, and they all exhibit excellent photocatalytic activity [1–4]. However, the photocatalysts only function with the light illumination. Once the light is removed, they will rapidly lose the activity. Therefore, developing new photocatalysts, which can work in the dark, is highly desirable.

Recently, it has been reported that some photocatalysts can store photoenergy under the light illumination, and then release it in the dark to stimulate the redox reaction. This phenomenon was called as post-illumination “memory” effect. Initially, Li et al. [5] fabricated TiON/PdO composites and found that, after pre-illumination for some time, the composites possessed strong disinfection capability in the dark. Further study showed that variable valence metal ions were the main factor for the “memory” activity [6]. Based on this, many photocatalysts with variable-valence metal ions have been developed [7,8], and they do present the “memory” activity. Very recently, high valence Bi-contained compounds such as NaBiO₃ [9] and BiO_{2-x} [10] have attracted much attention due to their excellent photoactivity. Considered the variable valence of Bi ions in the compounds, we conceive that

NaBiO₃/BiO_{2-x} composites may provide the excellent “memory” activity.

Herein, NaBiO₃/BiO_{2-x} composites were synthesized by a hydrothermal method. The photocatalytic activity and the “memory” activity were investigated by the degradation of organic pollutants under the light irradiation and in the dark, whose mechanism was discussed.

2. Experimental

NaBiO₃/BiO_{2-x} composites were synthesized by a hydrothermal method. The detailed preparation process, characterization, and relative tests were described in the [supplementary materials](#).

3. Results and discussion

The crystal structures of the samples were investigated by XRD (Fig. 1a). For BiO_{2-x}, all the diffraction peaks match well with the cubic phase BiO_{2-x} (JCPDS NO. 47-1057), and no other impurity peaks can be detected. Comparatively, NaBiO₃/BiO_{2-x} composites exhibit two sets of XRD peaks. The peaks with $2\theta = 28.2^\circ$, 32.6° , 46.9° , 55.6° , 58.3° and 68.5° can be indexed as (1 1 1), (2 0 0), (2 2 0), (3 1 1), (2 2 2) and (4 0 0) crystal planes of BiO_{2-x}, while the others can all be attributed to NaBiO₃, indicating the coexistence of NaBiO₃ and BiO_{2-x}. Fig. 1b shows the FT-IR spectra of the samples. As is expected, the main bands of NaBiO₃ and BiO_{2-x} can both be detected in NaBiO₃/BiO_{2-x}. The XPS survey spectra are shown in Fig. 1c. BiO_{2-x} consists of Bi and O element, while

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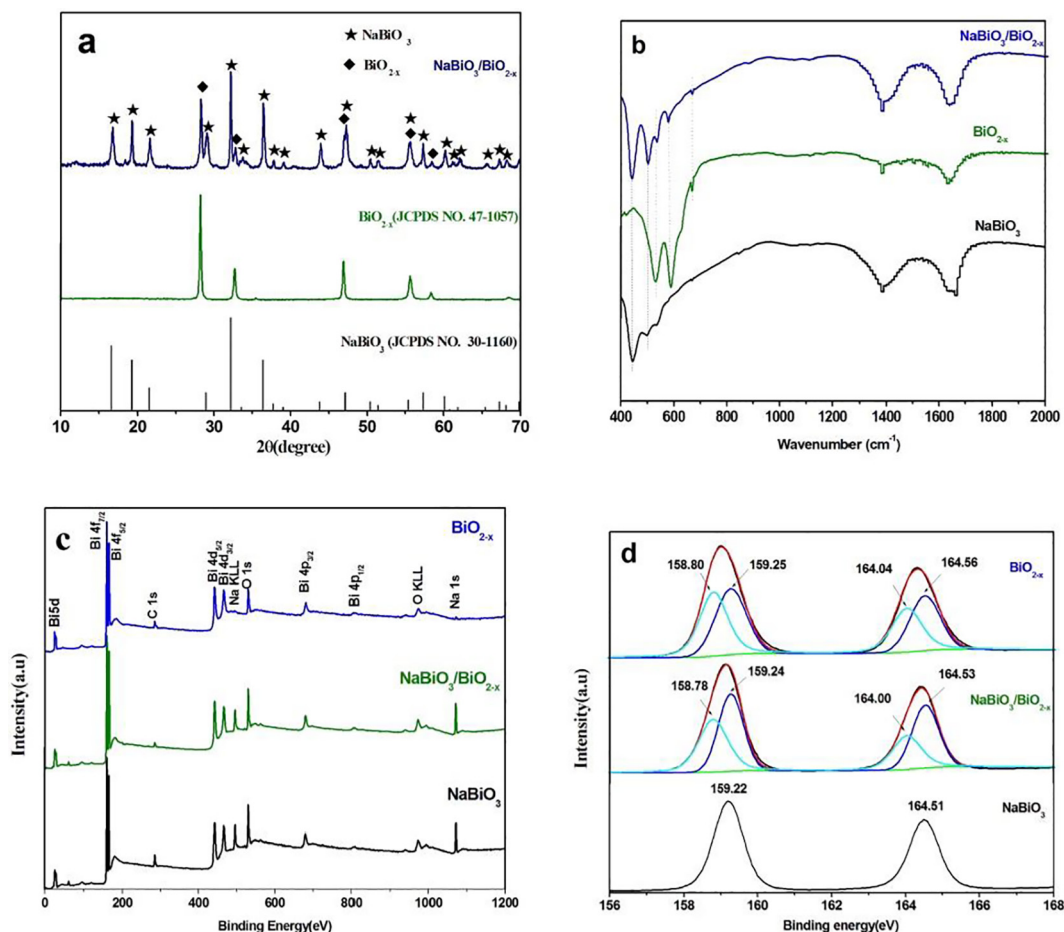


Fig. 1. (a) XRD pattern, (b) FTIR spectra and XPS spectra of (c) survey and (d) Bi4f for NaBiO₃, BiO_{2-x} and NaBiO₃/BiO_{2-x} composites.

NaBiO₃/BiO_{2-x} composites contain Bi, O, and Na element. The high resolution Bi 4f spectra of NaBiO₃ are located at 164.51 and 159.22 eV (Fig. 1d), referring to the normal state of Bi⁵⁺ [10]. Notably, these two peaks are both asymmetric for BiO_{2-x} and NaBiO₃/BiO_{2-x} with a shoulder at lower binding energies, which can be de-convoluted into two new bimodal peaks. The peaks at 164.56 eV and 159.25 eV are ascribed to Bi⁵⁺ species, while the peaks at 164.00 and 158.78 eV can be related to Bi³⁺ [11]. The Bi³⁺/Bi⁵⁺ atomic ratios in BiO_{2-x} and NaBiO₃/BiO_{2-x} are determined to be 0.89 and 0.67, respectively, which also indirectly confirms the coexistence of NaBiO₃ and BiO_{2-x}.

The morphologies of the samples were investigated by SEM and TEM. Commercial NaBiO₃ shows flower-like aggregates composed of irregular nanosheets (Fig. 2a), while BiO_{2-x} presents typical hierarchical architectures, which are constructed by regular nanosheets in a specific rule (Fig. 2b). By contrast, NaBiO₃/BiO_{2-x} composites consist of two kinds of nanosheets with different sizes (Fig. 2c). The large nanosheets cross with each other to form loose opening structure, in which the small ones are evenly dispersed. TEM image (Fig. 2d) shows the large nanosheets possess a particular shape with adjacent edge angle of approximately 120°, which is the characteristic of BiO_{2-x} [10]. The small ones take on rounded shape and are firmly adhered to the large ones, which can be related to the dehydrated NaBiO₃·xH₂O.

Fig. 3a shows UV–vis diffuse reflectance spectra of as-obtained samples. All samples present absorption in the visible light region, and the absorption edges are 480, 510 and 730 nm for BiO_{2-x}, NaBiO₃/BiO_{2-x} and NaBiO₃, respectively. The photocatalytic

activity of the samples was evaluated by the degradation of methyl orange(MO) under visible light irradiation. As shown in Fig. 3b, the photolysis of MO is negligible. BiO_{2-x} displays a weak degradation capability, over which 81.1% of MO is removed after 60 min of irradiation. Commercial NaBiO₃ demonstrates a better activity, which removes 91.2% of MO. By comparison, NaBiO₃/BiO_{2-x} degrade 97.7% of MO in the same time, suggesting the best photocatalytic performance. Besides, the composites also exhibit a superior activity toward the degradation of phenol (Fig. S1) and excellent stability. There is no obvious difference in the XRD patterns for NaBiO₃/BiO_{2-x} before and after photocatalysis (Fig. S2). The enhanced property can be ascribed to the heterojunction effect between NaBiO₃ and BiO_{2-x}, by which photo-generated electron-hole pairs are effectively separated just like BiOBr/TiO₂ [12]. Interestingly, after as-obtained samples are pre-illuminated for 30 min, the composites can still remove organic pollutants in the dark, and the MO removal rates are 21.5%, 97.6% and 37.2% for BiO_{2-x}, NaBiO₃/BiO_{2-x} and NaBiO₃, respectively (Fig. 3c). Obviously, NaBiO₃/BiO_{2-x} composites possess the highest post-illumination “memory” activity. Moreover, the “memory” activity can maintain for a long time. As shown in Fig. 3d, even if the pre-illuminated composites are stored in the dark for 7 h, they can still remove 50.3% of MO in the dark, which makes it possible for NaBiO₃/BiO_{2-x} to function all day.

Generally speaking, the post-illumination “memory” activity often involves a storing and release process of electrons [6,8]. To understand it, the open circuit potential change was recorded for as-obtained samples in response to light on/off. As shown in

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