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Carbon-modified bismuth titanate with an enhanced photocatalytic activity under nature sunlight

Jian Chen^a, Weigang Mei^a, Chao Liu^c, Chenhui Hu^b, Qianjing Huang^a, Ningna Chen^b,
Jing Chen^{a,*}, Rong Zhang^a, Wenhua Hou^{b,*}

^a College of Chemistry and Molecular Engineering, Nanjing Tech University, Nanjing 211816, PR China

^b Key Laboratory of Mesoscopic Chemistry of MOE, School of Chemistry and Chemical Engineering, Nanjing University, Nanjing 210023, PR China

^c School of Materials Engineering, Yancheng Institute of Technology, Yancheng 224051, PR China

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ABSTRACT

A novel carbon-modified bismuth titanate $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ (C/BTO) nanocomposites were successfully synthesized by a facile co-precipitation method. The resulted samples were characterized by X-ray diffraction (XRD), Fourier transform infrared (FT-IR) spectroscopy, scanning electron microscopy (SEM), X-ray photoelectron spectroscopic (XPS), N_2 adsorption–desorption and UV–vis diffuse reflectance spectroscopy (UV–vis DRS). The photocatalytic activities of the samples were measured through the photocatalytic degradation of methyl orange (MO) aqueous solution under sunlight. Compared with commercial P25, the resulted C/BTO showed an excellent photocatalytic activity. The mechanism for the improved photocatalytic activity was finally proposed.

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1. Introduction

As a new water treatment method, the photocatalytic technology has attracted special attention due to its potential application in the degradation of organic wastewater [1]. With the properties of stability, nontoxicity, low cost, high photocatalytic activity and so on, TiO_2 as a photocatalyst has been most widely investigated. However, the wide gap (3.0–3.2 eV) makes TiO_2 only be excited under UV light irradiation that is about 4% of solar light, which limits its practical application. To take the advantage of the visible light which is the most composition of solar energy (nearly 45%), exploring new visible light catalysts has become a focus in the photocatalytic field [2–4].

Recently, the novel Bi-based semiconductor materials have drawn greater concern for their high photocatalytic activity [5]. The hybridized 6s² of Bi^{3+} and O 2p make a new valence band which can reduce the band gap, therefore, most of Bi^{3+} – containing semiconductor compounds have a narrow band gap and good photocatalytic activity [6]. Bismuth titanate ($\text{Bi}_4\text{Ti}_3\text{O}_{12}$, abbreviated as BTO), a kind of layered perovskite compounds, is composed of alternatively stacking of $(\text{Bi}_2\text{O}_2)^{2+}$ layers and perovskite-like $(\text{Bi}_2\text{Ti}_3\text{O}_{10})^{2-}$ blocks along the c axis. It has been

widely explored for potential applications in optical displays, ferroelectric, and optoelectronic devices [7]. In recent years, $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ has been found to possess the ability of degrading organic pollutants and splitting water [8,9]. In addition, it has a high photocatalytic activity under visible-light [7]. Many fabrication techniques of crystalline $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ have been reported, such as chemical solution decomposition, solid-state reaction, hydrothermal synthesis, and so on [10]. However, $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ exhibits a low capability for the separation of electron–hole pairs and the photocatalytic activity of pure $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ is still far from practical application [11].

Studies have shown that carbon with the wide visible light absorption at a wavelength of 400–800 nm can enhance the photocatalytic activity and its high adsorption of organic pollutants can facilitate the interface reaction of photocatalysis [12]. For example, Ouyang et al. synthesized carbon doped ZnO composites with a porous structure which exhibited an excellent photocatalytic activity of RhB under solar-light irradiation [13]. Xiong et al. reported an *in situ* synthesis of C-doped $(\text{BiO})_2\text{CO}_3$ hierarchical microspheres which exhibited an enhanced visible light photocatalytic activity toward the removal of NO compared with the undoped $(\text{BiO})_2\text{CO}_3$, C-doped TiO_2 and N-doped $(\text{BiO})_2\text{CO}_3$ [14]. Using hydrothermal and calcination method, Li et al. synthesized C- Bi_2WO_6 which could totally decompose RhB in 4 h under visible light [15].

* Corresponding authors.

E-mail addresses: cj1908@126.com (J. Chen), whou@nju.edu.cn (W. Hou).

Herein, we report a facile synthesis of C/BTO and its photocatalytic activity for the degradation of MO under nature sunlight. The structure, morphology, and other characteristics are investigated in details. In addition, the degradation mechanism is also discussed.

2. Experimental

All the chemicals were of analytical grade and used as received without further purification. Detailed synthesis process, characterization means and photocatalytic evaluation were given in the [Supplementary information \(SI\)](#).

3. Results and discussion

The layered structure and crystallinity of the resulting samples (BTO, H⁺/BTO and C/BTO) were characterized by XRD. As shown in [Fig. 1a](#), the XRD pattern of the as-prepared BTO matches well with the published data (JCPDS no. 35-0795) and the layered structure of BTO is clearly visible ([Fig. 1c](#)). After acidification and C modification, the characteristic (060) and (080) diffraction peaks of layered structure are disappeared, while the intensity of other main peaks is significantly reduced, indicating that the original ordered layered structure has been destroyed. From SEM image in [Fig. 1d](#), we can see that C/BTO has no obvious ordered layered structure. [Fig. 1b](#) displays the FT-IR spectra of BTO, H⁺/BTO and C/BTO. The bands at 790 cm⁻¹ and 658 cm⁻¹ in BTO are attributed

to the stretching vibrations of Bi-O and Ti-O [6]. After acidification and C modification, the intensity and peak width of these two peaks are changed, but the basic characteristics are remained. It demonstrates that the basic structure is not changed significantly, being consistent with the XRD results. The band at 1380 cm⁻¹ in H⁺/BTO is attributed to the vibration of NO bond which may be derived from the surface adsorbed NO₃⁻. The peaks located at ~1633 and 1693 cm⁻¹ may belong to the characteristic absorptions of “C=O” [16], while the band at 1425 cm⁻¹ is ascribed to “-COO⁻” [17]. Moreover, the surface chemical composition of C/BTO was also investigated by XPS analysis ([Fig. S1](#)). The survey spectrum shows the presence of Bi, Ti, O and C without other elements detected, and the C 1s spectrum shows two peaks. The peak at 284.2 eV may be assigned to the C-C bond with sp² orbital, while the peak at 288.1 eV belongs to C=O bond of carbonate species [17]. All these evidence that C modification is successful. Accordingly, we suspect that the ethanol was oxidized by nitric acid to be carbonyl-containing species coated onto the surfaces of BTO.

The nitrogen adsorption-desorption isotherms are shown in [Fig. 2a](#). Both H⁺/BTO and C/BTO present an obvious H3 hysteresis loop, indicating the presence of slit-like pore structure due to the stacking of plate-like particles [18]. By comparison, BTO has no apparent hysteresis loop, revealing that the sample is mainly non-porous. The BET surface area of BTO is only 8.9 m²/g. After acidification and C modification, the BET surface areas of H⁺/BTO and C/BTO have greatly increased to 113.3 and 107.9 m²/g, respectively.

The optical absorbance of the obtained samples was measured by UV-visible diffuse reflectance spectra. As shown in [Fig. 2b](#), BTO

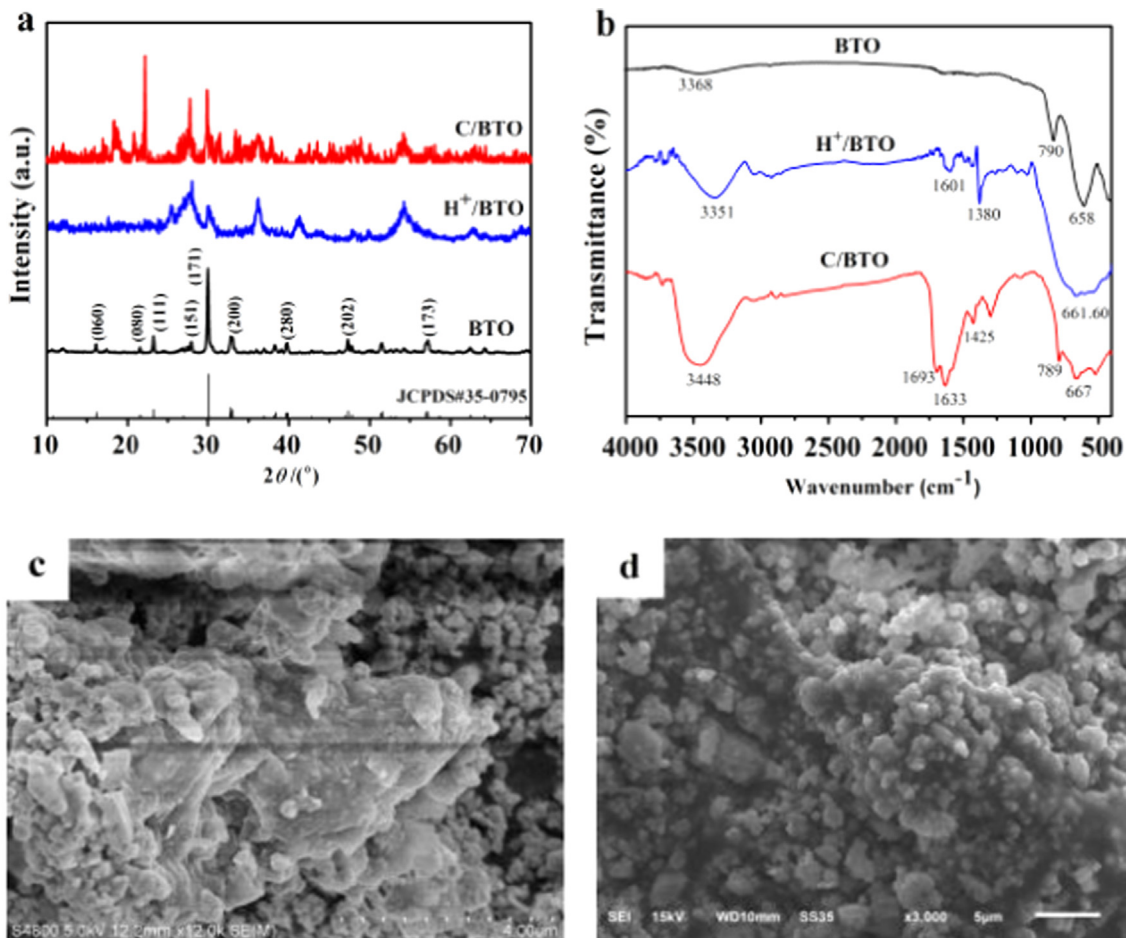


Fig. 1. XRD patterns (a) and FTIR spectra (b) of the relevant samples; SEM images of BTO (c) and C/BTO (d).

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