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Synthesis of AgI/Bi₂MoO₆ heterojunctions and their photoactivity enhancement driven by visible light



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ARTICLE INFO

Article history:

Received 16 January 2016

Received in revised form

16 March 2016

Accepted 23 March 2016

Available online 25 March 2016

Keywords:

Deposition-precipitation

AgI/Bi₂MoO₆ composites

Photocatalysis

ABSTRACT

AgI/Bi₂MoO₆ heterojunction photocatalysts were successfully synthesized by deposition-precipitation of AgI nanoparticles on Bi₂MoO₆ nanoplates. The as-synthesized photocatalysts were characterized by XRD, TEM and XPS. Effect of AgI loaded on photoactivity of AgI/Bi₂MoO₆ heterojunctions was investigated through the photodegradation of rhodamine B (RhB) as a model toxic contaminant under visible light. The 10 wt% AgI/Bi₂MoO₆ heterojunctions have the highest RhB removal efficiency of 92.89% within 40 min.

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1. Introduction

Visible driven photocatalysts have attracted worldwide researchers because they can apply for splitting of water and the degradation of organic pollutants by approximately 43% visible light of solar radiation [1–3]. Bi₂MoO₆ visible driven photocatalyst with 2.9 eV is a typical Aurivillius phase containing perovskite layers (Bi₂O₂)(A_{m-1}B_mO_{3m+1}) [4–6]. A number of researchers have studied photocatalytic activities of Bi₂MoO₆ to decompose organic contaminants under visible light [1,3–6]. Moreover, photocatalytic activities of Bi₂MoO₆ are limited by rapid recombination of photoexcited carriers [1,2]. Thus, effective visible-light-driven photocatalyst was developed to have a great benefit for environmental treatment. Heterojunction photocatalysts can improve the efficiency of visible-light-driven photocatalyst because they can reduce the recombination rate of photoinduced electron-hole pairs [2]. As one of the excellent sensitizers, AgI can be used to modify wide-band-gap semiconductors to enhance the visible absorption ability and to utilize solar energy due to its narrow band gap of ~2.8 eV [7].

In this work, AgI/Bi₂MoO₆ heterojunctions were successfully

synthesized by deposition-precipitation at room temperature. The AgI/Bi₂MoO₆ heterojunctions showed high photocatalytic activity to decompose rhodamine B (RhB) under visible light.

2. Experiment

Each of 5 mmol Bi(NO₃)₃ · 5H₂O and Na₂MoO₆ was dissolved in each 100 ml RO water and mixed together. The obtained solution was stirred and adjusted the pH to 6 by 3 M NaOH solution. The mixture was transferred to a 200 ml Teflon-lined stainless steel autoclave, tightly closed and maintained at 180 °C in an electric oven for 20 h. The obtained precipitates were washed with distilled water and ethanol, dried at 80 °C for 24 h and collected for the synthesis of heterostructure nanocomposites.

To synthesize heterostructure AgI/Bi₂MoO₆ nanocomposites, different concentrations of 0–10% AgNO₃ and NaI by weight and 2.5 g Bi₂MoO₆ sample were dissolved in 100 ml RO water under magnetic stirring for 24 h. Subsequently, the products were separated by filtering, washed with ethanol and dried at 80 °C in an electric oven for 24 h for further characterization and photocatalytic testing.

3. Results and discussion

Fig. 1a shows XRD patterns of pure Bi₂MoO₆ and 1, 5 and

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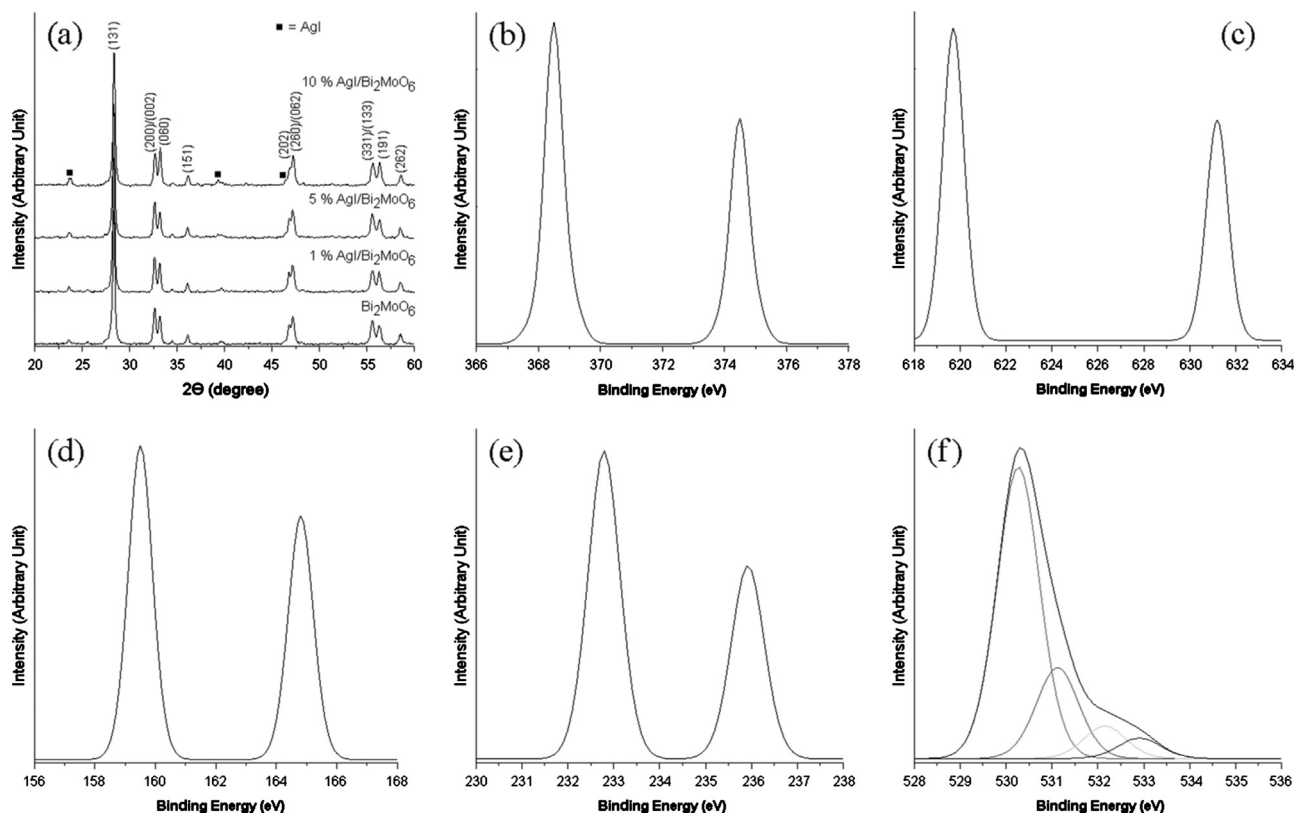


Fig. 1. (a) XRD patterns of 0–10 wt% AgI/Bi₂MoO₆ heterojunctions and (b–f) XPS spectra of Ag3d, I3d, Bi4f, Mo3d and O1s of 10 wt% AgI/Bi₂MoO₆ heterojunctions, respectively.

10 wt% AgI/Bi₂MoO₆ heterojunctions. For the pure sample, it can be indexed to orthorhombic Bi₂MoO₆ phase (JCPDS no. 21-0102 [8]) without impurity detection. For the analysis of AgI/Bi₂MoO₆ heterojunctions, additional hexagonal AgI phase (JCPDS no. 09-0374 [8]) was detected, implying the coexistence of the two phases. Diffraction peaks of the composites are clearly related to AgI and Bi₂MoO₆ phases and the loaded AgI of the composites did not change Bi₂MoO₆ crystalline phase. The chemical state of 10 wt% AgI/Bi₂MoO₆ heterojunctions (Fig. 1b–f) shows XPS peaks at 374.48 and 368.49 eV, ascribed to Ag3d_{3/2} and Ag3d_{5/2} of Ag⁺ [7,9], those at 619.70 and 631.20 eV are for I3d_{5/2} and I3d_{3/2} [7], including two strong peaks at 164.80 and 159.51 eV are assigned to Bi4f_{5/2} and Bi4f_{7/2} [7,9,10], respectively. The analysis confirms the presence of Bi³⁺ cations containing in the AgI/Bi₂MoO₆ composites. Two peaks of Mo3d_{5/2} at 232.81 eV and Mo3d_{3/2} at 235.91 eV were assigned to Mo⁶⁺ species [9,10]. The O1s XPS spectrum can be deconvoluted into four peaks relating to Bi–O bond at 530.25 eV, Mo–O bond at 531.10 eV, surface adsorbed O species at 532.13 eV and C–O bond of adsorbed ambient air at 532.89 eV [7,9,11]. The above results revealed the successful synthesis of AgI/Bi₂MoO₆ heterojunctions.

TEM and HRTEM images of Bi₂MoO₆ and AgI/Bi₂MoO₆ heterojunctions are shown in Fig. 2. The as-synthesized Bi₂MoO₆ was composed of nanoplates with edge length of 100–200 nm. Their surfaces were very smooth with no other particles on top. The SAED pattern (inserted of Fig. 2a) of a nanoplate, indexed to the (060), (062) and (002) planes with electron beam along the [100] direction, revealed the presence of single crystal with orthorhombic Bi₂MoO₆ structure. TEM images of 5 and 10 wt% AgI/Bi₂MoO₆ samples showed that Bi₂MoO₆ remained as nanoplates with AgI nanoparticles on top. The amount of AgI nanoparticles was increased with the increase of AgI content. Moreover, AgI nanoparticles with < 10 nm fully covered on Bi₂MoO₆ surface of

10 wt% AgI/Bi₂MoO₆. HRTEM image of 10 wt% AgI/Bi₂MoO₆ heterostructures revealed the interplanar space of 0.373 nm corresponding to the (002) crystallographic plane of AgI, indicating the formation of AgI/Bi₂MoO₆ heterostructure. The as-synthesized heterostructure has better charge separation and more efficient carrier transfer comparing to pure Bi₂MoO₆ nanoplates. Thus their photocatalytic activities were improved [2,9].

Photocatalytic activities of the as-synthesized samples were investigated by photodegradation of RhB dye as a model reaction in water to demonstrate the degradation ability of the photocatalysts. Fig. 3a shows UV–visible absorption of 200 ml aqueous solution containing 1 × 10^{−5} M RhB and 20 mg photocatalyst under visible light for different lengths of time. It can be seen that the intensity of RhB at λ_{max} of 554 nm gradually decreased as the exposure time was lengthened, indicating the degradation of RhB. Moreover, the absorption peak shifted from 554 nm to 498 nm, corresponding to the color change of the solution from pink to cyan. The gradual hypsochromic shift of the absorption peak to a shorter wavelength is caused by the N-deethylation of RhB during irradiation [3,6,7]. This hypsochromic shift was attributed to degrade in sequence: deethylation of RhB to yield N,N,N′-triethyl rhodamine (TER, 539 nm), N,N′-diethyl rhodamine (DER, 522 nm), N-ethyl rhodamine (MER, 510 nm) and rhodamine at 498 nm [12]. A proposed mechanism of RhB degradation by the AgI/Bi₂MoO₆ heterostructures can be explained by the following. Band energy levels of AgI and Bi₂MoO₆ were illustrated in the previous reports [13–15]. Under visible light, both AgI and Bi₂MoO₆ are excited by visible light to produce the electron (e[−]) in conduction band (CB) and hole (h⁺) in valence band (VB). In this case, electrons can easily diffuse from the CB of AgI to CB of Bi₂MoO₆ because CB of AgI (−0.43 V w.r.t. NHE) is more negative than CB of Bi₂MoO₆ (−0.32 V w.r.t. NHE). Concurrently, holes can diffuse from VB of Bi₂MoO₆ to VB of AgI because VB of AgI (2.61 V w.r.t. NHE) is more

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