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Improved photocatalytic activity and durability of AgTaO₃/AgBr heterojunction: The relevance of phase and electronic structure

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ABSTRACT

AgTaO₃/AgBr heterojunction was constructed for visible light driven photocatalytic purpose in order to investigate the relevance of phase conversion, electronic structure and photocatalytic properties. The result indicated that AgBr grafted on AgTaO₃ to form AgTaO₃/AgBr heterojunction gave intense visible light absorption, which exhibits highly enhanced photocatalytic performance than their individual counterpart. Theoretical and experimental investigation showed that the matched electronic structure between AgTaO₃ and AgBr induced an efficient transfer of photogenerated electrons from AgBr to AgTaO₃, leading to efficient charge separation and the subsequent improved photocatalytic activity. Partial AgBr converted to AgBr/Ag during the photocatalytic process, leading to the construction of ternary AgTaO₃/AgBr/Ag photocatalyst. Because of the surface plasmon resonance effect of Ag, the resulting AgTaO₃/AgBr/Ag exhibited wide range absorption and improved charge separation efficiency, which showed high durability and superior photocatalytic activity toward methyl orange degradation. On the basis of spin resonance measurement and trapping experiment, it is expected that photogenerated electrons, O₂^{•-}, and OH[•] active species dominate the photodegradation of methyl orange.

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1. Introduction

Photocatalysis based on semiconductors harnesses considerable research attentions due to its potential applications in environmental remediation, hydrogen production by water splitting and chemical conversion for valuable products [1–5]. Till now, a variety of semiconductors for efficient photocatalytic purpose have been fully investigated. Among them, tantalate-based semiconductors are considered to be excellent photocatalysts due to their strong driving force for photoredox reduction, high chemical and physical stability, and superior photocatalytic activity [6–10]. However, stronger photoredox driving force is often accompanied with a wider band gap that considerably limits visible light absorption. In this regard, the improvement of solar light absorption of tantalate-based semiconductors for efficient visible light driven photocatalytic performance is still necessary.

Numerous methodologies have been adopted to tailor the electronic structure for improving the visible light response of

tantalate-based semiconductors by metal and/or nonmetal doping, heterojunction and surface/interface engineering [11–14]. In particular, the heterojunction a tantalate-based semiconductor with narrow band gap semiconductors can effectively improve the photocatalytic activity compared with the single component counterpart [15]. The enhancement of the photocatalytic activity is thought to be attributed to the efficient separation of photogenerated charge carriers as a consequence of the matching electronic structure such as band gap energy and the potential energy levels of valence band and conduction band. Recently, the enhanced visible light driven photocatalytic activity has been achieved in several heterojunction photocatalysts, including g-C₃N₄/NaTaO₃, TaON/Bi₂O₃, carbon-Ta₂O₅, Sr₂Ta₂O_{7-x}N_x/graphene and so on [16–19], in which tantalate-based semiconductors play critical roles. In order to develop tantalate-based heterojunction systems, it is still important to explore the phase, electronic structure and the underlying mechanism for efficient visible light absorption and photocatalytic activity.

Silver tantalate (AgTaO₃) exhibits distorted pseudocubic structure with a perovskite structure has started to obtain surged research attention due to its promising applications as solid lubricant, dielectric material and photocatalyst [20]. As previously reported, the band gap of AgTaO₃ is determined to be about 3.4 eV [21], which is only reactive under ultraviolet (UV) light irradiation,

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limiting its solar energy applications. A systematic experimental identification of AgTaO_3 and its relevant heterojunctions by modulating phase structure, chemical composition as well as interfacial contact is advantageous for tailoring the photophysical and photochemical properties of AgTaO_3 as well as other isostructural semiconductors, such as NaTaO_3 , AgNbO_3 , CuTaO_3 and so on. Most recently, much research attention has been focused on silver halide AgX ($X = \text{Cl}, \text{Br}, \text{I}$) because of their high photocatalytic performance toward the removal of deleterious contaminant [22–24]. Unfortunately, silver halide often suffers from chemical instability under visible light irradiation, being of a hindrance to its potential applications [25]. Recent studies suggests that heterojunction of AgBr can with other semiconductors can effectively improve the photocatalytic activity and stability of AgBr [26–28]. Herein, with respect to the above-mentioned problems, we purposed to develop $\text{AgTaO}_3/\text{AgBr}$ composite for efficient and renewable visible light photocatalyst by modulating the phase structure, chemical composition as well as the electronic structure, which is helpful to identify the underlying mechanism of the heterojunctions.

2. Experimental section

2.1. Procedure for the synthesis of the $\text{AgTaO}_3/\text{AgBr}$ composite photocatalysts

2.1.1. Synthesis of AgTaO_3

AgTaO_3 sample was synthesized by a solid state reaction. Ta_2O_5 was mixed with excess amounts (2–5 mol%) of Ag_2O in an agate mortar. The mixture was calcined in an alumina crucible at 1050°C for 5 h. The obtained sample was treated with certain amount of nitric acid to remove the excess silver and washed with distilled water for several times, and then dried in an oven at 80°C for 10 h.

2.1.2. Synthesis of AgBr

Pure AgBr was prepared via a solution method. Briefly, AgNO_3 and NaBr solutions with Ag/Br molar ratio of 1:1 were mixed to form a yellow suspension. After aged for 2 h, the suspension was filtered and washed several times using distilled water, then dried at 80°C for 10 h.

2.1.3. Synthesis of $\text{AgBr}/\text{AgTaO}_3$

All the following processes were carried out in a dark situation. Briefly, 0.6 g of AgTaO_3 was added to 60 mL of distilled water to form a white suspension. The suspension was sonicated for 30 min at room temperature. 1.3 g of KBr was added to the above suspension with magnetically stirring for 2 h. Then, 0.6 g poly(ethyleneglycol)-block-poly(ethylene glycol) (P123) was added to the suspension and stirred for 1 h. Subsequently, given amount of AgNO_3 dissolved in 1.8 mL of $\text{NH}_3 \cdot \text{H}_2\text{O}$ (containing 25 wt% of NH_3) was added to the above mixture. The initial weight ratio of $\text{AgBr}/(\text{AgTaO}_3 + \text{AgBr})$ were modulated to be 30 wt%, 50 wt%, 70 wt%, 90 wt%, and defined as AA-30, AA-50, AA-70, AA-90, respectively. The resulting suspensions were stirred at room temperature for 12 h. The as-resulted products were filtered, washed and dried at 80°C over night.

2.2. Characterization of photocatalysts

X-ray power diffraction (XRD) was used to characterize the phase structure of the as-prepared samples on a Rigaku DMAX2500 X-ray diffractometer using a copper target. Transmission electron microscopy (TEM) was performed on a JEM-2010 apparatus with an acceleration voltage of 200 kV. Energy-dispersive spectroscopy (EDX) data were obtained using scanning electron microscopy (SEM) on a S-4800 apparatus working at 10 kV. Optical diffuse reflectance spectrum was measured using a Lambda 750 s spectrometer. XPS analyses were performed on the ESCALab220i-XL

with a monochromatic $\text{Al K}\alpha$ and charge neutralizer. The surface photovoltage measurement system is constructed by a sample chamber, a lock-in amplifier with a light chopper and a source of monochromatic light (provided by a 500 W xenon lamp) and a monochromator. The monochromator and the lock-in amplifier were equipped with a computer. The analyzed product is assembled as a sandwich-like structure of ITO-product-ITO (ITO is an indium tin oxide electrode). The presence of hydroxyl radicals and superoxide radicals was measured by EPR spectra, which was performed on an ER200-SRC electron spin resonance spectrometer (Bruker, Germany) at 3186 G and 9056.895 MHz.

2.3. Photocatalytic activity tests

The photocatalytic activity of the as-prepared samples was evaluated by using methyl orange as the probe molecule under visible light irradiation at room temperature. In brief, 50 mg of the sample was placed in a 100 mL beaker containing 50 mL 2×10^{-5} M methyl orange solution to form a suspension. Before illumination, the above suspension was stirred in dark in order to establish the absorption–desorption equilibrium of methyl orange on sample surfaces. Then, the suspension was irradiated by a 300 W Hg lamp using a filter ($\lambda \geq 420$ nm) as a cutoff, which is about 10 cm away from the beaker. Then, 5 mL of the suspension was extracted at given intervals and centrifuged at a rate of 8000 rpm for 10 min. UV–vis absorption spectra of the supernatant was measured using a Lambda 750 s Spectrometer.

3. Results and discussion

XRD technique was adopted to investigate the crystallographic structure of the as-prepared samples. Fig. 1 shows the XRD patterns of AgTaO_3 , AgBr and their heterojunctions. As for AgTaO_3 , all diffraction peaks can be well indexed to perovskite-type phase of AgTaO_3 (JCPDS card no. 72-1383). No trace of additional diffraction peaks was observed, suggesting a single phase perovskite structure is formed, being similar to Xu's results [29]. By using the Rietica Rietveld program based on a least-squares method, the lattice parameter of AgTaO_3 was estimated to be $a = 5.523 \text{ \AA}$, and $c = 13.623 \text{ \AA}$, being close to the standard data of AgTaO_3 . The narrowed diffraction peaks suggest high crystallinity and fine nature of AgTaO_3 . Besides AgTaO_3 , the XRD pattern of AgBr is also given for comparison. As shown in Fig. 1, all narrowed diffraction peaks of AgBr can be ascribed to the typical cubic structure, which is identical to the standard data of AgBr (JCPDS card no. 06-0438). As AgTaO_3

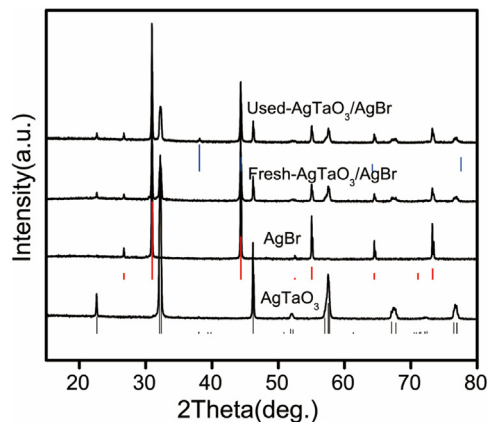


Fig. 1. XRD patterns of the as-prepared samples. Vertical bars represent the standard diffraction data from the JCPDS files for AgTaO_3 (72–1398, black line), AgBr (06-0438, red line) and Ag (87-0597, blue line). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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