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A novel method for the synthesis of $Ag_3VO_4/Ag_2VO_2PO_4$ heterojunction photocatalysts with improved visible-light photocatalytic properties

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ABSTRACT

$Ag_3VO_4/Ag_2VO_2PO_4$ heterostructure with highly enhanced visible light photocatalytic activity were synthesized by using $Ag_2VO_2PO_4$ as the sacrificed template in different concentration of ammonia solution. Their crystalline structure, morphology, optical, and electrochemical properties were characterized by X-ray diffraction, scanning electron microscopy, X-ray photoelectron spectroscopy, UV–vis diffuse reflectance spectroscopy, infrared spectroscopy, and electrochemical measurements. The photocatalytic degradation activities of as-prepared samples were evaluated by the photocatalytic of methylene blue (MB), methyl orange (MO) and imidacloprid in the aqueous phase. Compared with bare $Ag_2VO_2PO_4$, the etched samples $Ag_3VO_4/Ag_2VO_2PO_4$ exhibit the significantly enhanced photocatalytic activity under visible light irradiation. The $Ag_3VO_4/Ag_2VO_2PO_4$ with 0.15 M etched sample exhibits the highest activity, which are nearly 3.8, 8.6 and 9 times higher than bare $Ag_2VO_2PO_4$ for removal of MB, MO and imidacloprid, respectively. The improved photocatalytic performance of $Ag_3VO_4/Ag_2VO_2PO_4$ can be attributed to the formation of $Ag_2VO_2PO_4/Ag_3VO_4$ heterojunction, which suppresses the recombination of photoinduced charges and prolongs the lifetime of the charges. Moreover, the photostability and dominant active species were investigated.

1. Introduction

Solar energy is one of the promising renewable energy sources in the world, due to its unique advantages, such as naturalness, free availability, non-polluting characteristics and inexhaustibility [1]. The effective harvest and utilization of solar energy has become an irresistible trend and is expected to address the energy shortage and environmental pollution caused by the quick industry development in the last decades [2,3]. Compared with adsorption [4,5], the semiconductor-based photocatalysis has been regarded as one promising technique to harvest and convert solar energy to chemical energy [6–8]. For half a century, material, chemical and physical scientists and engineers have been working hard to find effective photocatalysts. Among them, TiO_2, ZnO [9–11] the earliest reported and classic photocatalyst, have attracted many attention due to its low price, nontoxicity, strong oxidation ability, and excellent photostability. However, TiO_2 with large band gap (~3.2 eV) can only respond to UV light and is incapable of absorption in visible light range, which greatly limits the efficiency of utilizing sunlight [12,13]. To overcome the disadvantage, tremendous efforts have been devoted to enhance its photocatalytic activity under visible light irradiation. For example, coupling with a narrow band gap semiconductor, ion doping, noble metal decoration, etc. On the other side, another strategy is to develop non-TiO_2-based visible-light active photocatalysts with narrow energy gaps and strong absorption of visible light, such as $NiFe_2O_4$, $BiVO_4$, BiOX (X = Br, Cl, I), Ag_3VO_4, Ag_2O , $AgNbO_3$, $Zn_{1-x}Cd_xS$, and g-C_3N_4 [14–25].

In recent years, Ag-based photocatalysts [26] including Ag_2O [15], Ag_3VO_4 [27], Ag_3PO_4 [28], Ag_2CO_3 [29], AgX (X = Br, Cl, I) [30,31], Ag_2CrO_4 [32] have attracted great interest as an effective materials making full use of solar energy, due to their appropriate band gaps and excellent visible-light response [33–35]. Despite all this, there are still disadvantages, that is, the recombination of photo-induced electron-hole ($e^- - h^+$) pairs and the self-corrosion, leading to the decreased photoactivity and the consumption of the photocatalysts. For this, great efforts have been devoted to the design and preparation of Ag-based photocatalysts with improved photocatalytic performance. In our previous work, we firstly applied $Ag_2VO_2PO_4$ as a new photocatalyst. Like most Ag-based materials, $Ag_2VO_2PO_4$ with a relative narrow band gap of ~2.20 eV can absorb some portion of visible light [36]. However, its photoactivity is still needed to be improved further through coupling with other semiconductors to construction of heterojunction, which has been proved to be a effective method.

Chemical etching of the solid in solution are widely used in photocatalyst preparation, which always involves the in-situ formation of

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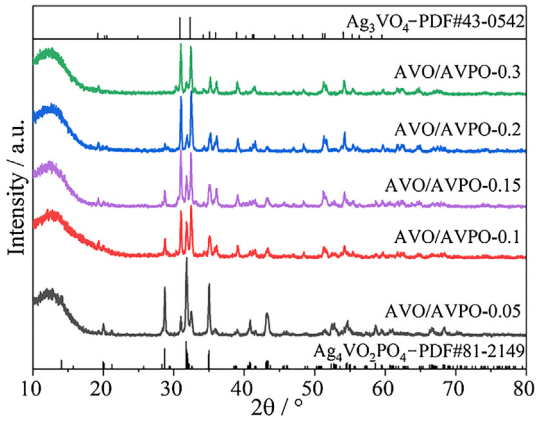

Fig. 1. XRD patterns of AVO/AVPO photocatalysts at different ammonia concentration.

new solid surface whose structure (composition and morphology) and subsequent reactivity are dynamically changed [37,38]. The solid particles exchange ion in liquid, and the surface, corn or edge were etched, thus, there must be a new nanocrystals and then the heterostructure is formed or various nanostructures, and exhibited much higher photocatalytic activities [39].

In this work, we etched $Ag_2VO_2PO_4$ using ammonia solution of different concentrations for the first time. Interestingly, Ag_3VO_4/$Ag_2VO_2PO_4$ composites with different morphologies and phase compositions were obtained. The Ag_3VO_4/$Ag_2VO_2PO_4$ heterojunction photocatalysts exhibit dramatically enhanced photoactivities than pristine $Ag_2VO_2PO_4$.

2. Experimental section

2.1. Synthesis of $Ag_2VO_2PO_4$ and Ag_3VO_4/$Ag_2VO_2PO_4$

The pristine $Ag_2VO_2PO_4$ was synthesized via a hydrothermal method at 230 °C for 4 days using the Ag_2O (1.236 g), NH_4VO_3 (0.6226 g), H_3PO_4 (2.4 mL, 85%) and deionized water (45.6 mL) as raw materials, which was reported in our previous work [36]. Ag_3VO_4/$Ag_2VO_2PO_4$ (AVO/AVPO) heterostructure photocatalysts were synthesized by using as-synthesized $Ag_2VO_2PO_4$ as the sacrificed template in ammonia solution of different concentrations. In the etching procedure, 0.5 g $Ag_2VO_2PO_4$ was dispersed in ammonia solution with different concentrations (0.05, 0.1, 0.15, 0.2 and 0.3 M), followed by 24 h of stirring. After collected and washed with distilled water and ethanol for several times, the powder was dried at 60 °C in air. For conveniently describing, we denote the AVO/AVPO as AVO/AVPO - 0.05, - 0.1, - 0.15, - 0.2 and - 0.3, respectively, to distinguish the different concentrations.

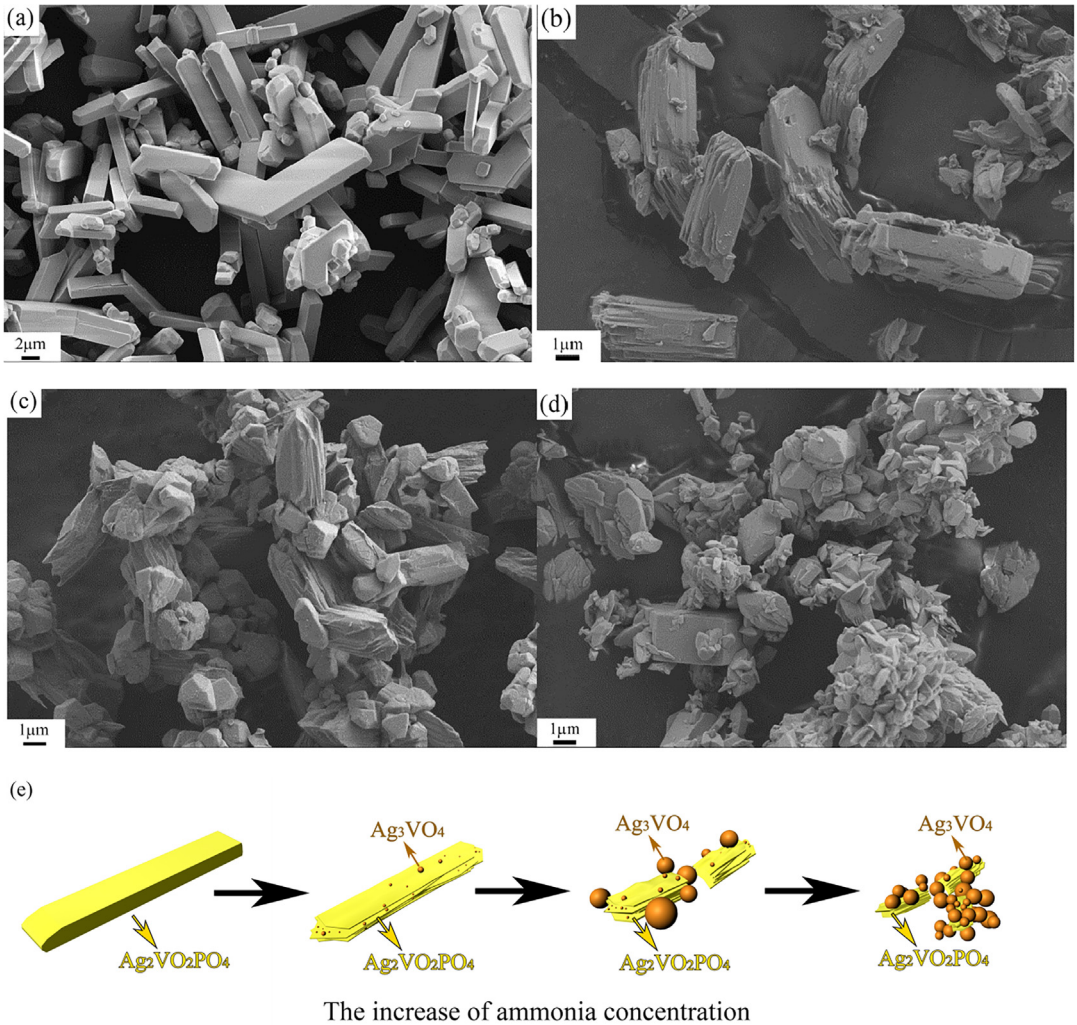

Fig. 2. SEM images of (a) bare $Ag_2VO_2PO_4$ and AVO/AVPO heterojunction photocatalysts (b) AVO/AVPO - 0.05, (c) AVO/AVPO - 0.15, and (d) AVO/AVPO - 0.2, respectively; (e) Schematic illustration of morphology changes for $Ag_2VO_2PO_4$ and AVO/AVPO - 0.05, - 0.15, - 0.2.