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Original Research Paper

One step pyridine-assisted synthesis of visible-light-driven photocatalyst Ag/$AgVO_3$

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ABSTRACT

Ag/$AgVO_3$ nanorods have been obtained for the first time by one step hydrothermal method with the contribution of pyridine. The prepared sample was systematically characterized. Their photocatalytic properties were investigated by degrading acid orange II (AO-II) under visible light. Compared with pure $AgVO_3$, Ag/$AgVO_3$ nanorods showed much higher photocatalytic activity and stability. The enhanced photocatalytic activity was attributed to Ag nanoparticles (NPs) with a strong surface plasmon resonance (SPR). Further studies indicate that the photogenerated holes (h^+) and superoxide radical anions ($\cdot O_2^-$) were major active species. Ag/$AgVO_3$ nanorods has a huge potential application in wastewater treatment.

1. Introduction

Photocatalysis has gained increasing attention from researchers because it is an ideal technology to solve the energy shortage and environmental problems [1–3]. For the past decade or more, various semiconductors, including ZnO [4,5], TiO_2 [6,7], CuO [8,9], have been extensively studied. However, most of them had a high rate of charge carrier recombination and almost no visible light absorption. Hence, many efforts have been devoted to address these two shortcomings by doping TiO_2, such as with nonmetals N-doping [10] and self doping [11], or decorating with some narrow-band-gap semiconductors such as SnO_2-decorated $ZnSn(OH)_6$ [12], $Bi_2Ti_2O_7/TiO_2$ [13]. To some extent, their photocatalytic properties have been improved by these methods. Nevertheless, their unsatisfactory activities and low efficiencies in the utilization of solar irradiation concentrated in visible light regions is difficult to meet their practical applications. Therefore, it is necessary to develop new and highly deficient visible light-active photocatalysts and effectively promote the separation of photogenerated electrons and holes. Up to now, a series of Ag-based photocatalysts have presented great abilities to photodegrade the organic pollutants under visible light irradiation. Among these new photocatalysts, silver vanadium oxide (SVO) ($AgVO_3$, $Ag_2V_4O_{11}$, Ag_3VO_4, etc.) have attracted enormous attention owing to their extensive application, unique hybridization of valence bands of V 3d, O 2p and Ag 4d orbits in SVOs, which could be utilized as visible-light-sensitive photocatalyst [14–17]. As a typical SVO compound, $AgVO_3$ with excellent optical absorption in the visible light region has attracted considerable attention due to its narrow band gap (ca. 2.3 eV), unique electronic structure and well crystallization. However, the catalytic activity of $AgVO_3$ is low due to its low capability of separating electron-hole pairs in the photocatalytic reaction. There are usually two ways to enhance its visible photocatalytic activity. One way is to design specific structures with different morphologies that lead to better performance. Among them, one-dimensional (1D) structure is expected to show higher activities owing to having a larger aspect ratio for extensive charge separation on their surfaces. Another way is to combine with plasmonic nanoparticles (NPs) made of noble metals (such as Au and Ag), which is endowed with high absorption coefficient in a broad visible spectral range due to their strong surface plasmon resonance (SPR) [18–20]. For instance, Wei [21] and co-workers have developed Ag/$AgVO_3$ nanoribbons plasmonic photocatalyst via in situ reduction of $AgVO_3$ by $NaBH_4$ at room temperature, which showed high photocatalytic activity. Malkhasian et al. [22] have reported Pt/$AgVO_3$ plasmonic photocatalyst via combining a hydrothermal method with photo-assisted deposition. The results reveal that Pt/$AgVO_3$ plasmonic photocatalyst have enhanced photocatalytic activity. Peng [23] and co-workers have prepared Ag/$AgVO_3$ 1D nanometer material, and the photocatalytic activity for the degradation of bisphenol A was investigated under visible light irradiation. The results showed that Ag/$AgVO_3$ exhibited higher photocatalytic activity than bare $AgVO_3$. Therefore, the construction of $AgVO_3$-based composite photocatalysts by introducing noble metals with SPR is an effective means to

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enhance their photocatalytic activity. Nevertheless, the preparation methods mentioned in the literature have been subjected to two steps or more.

Herein, we demonstrate one step hydrothermal method fabricating 1D $Ag/AgVO_3$ nanorods with pyridine (Py). The pyridine which was introduced to the synthesis system, not only works as a coordination agent of Ag^+, but also as a reductant for reducing Ag^+ ions to metal Ag. The as-prepared $Ag/AgVO_3$ catalyst was characterized by scanning electron microscopy, transmission electron microscope, X-ray diffraction, Raman spectrums, X-ray photoelectron spectroscopy, and UV–Vis diffuse reflectance spectra. Photocatalytic activity and recycling performance were investigated for degradation of AO-II under visible light irradiation.

2. Experimental

2.1. Catalysts preparation

All chemicals were analytical grade and obtained from Aladdin Reagents Company. $Ag/AgVO_3$ were prepared by one step hydrothermal method with pyridine. Typical synthesis was described as the following. Firstly, 30 mL solution of 1.25 mmol $AgNO_3$ and 3 ml pyridine were mixed together in a 100 mL Teflon-lined. Then 30 mL equimolar amounts of NH_4VO_3 solution was added with drop by drop to the above solution under vigorous stirring. After ultrasonic treatment for 30 min, the autoclave was sealed and maintained at 180 °C for 24 h, and then cooled to room temperature naturally. The products were collected by centrifugation, and alternately washed with distilled water and absolute ethanol several times, and finally dried in a vacuum at 70 °C in the dark. For the sake of contrast, $AgVO_3$ were prepared under the same condition except for no addition of pyridine.

2.2. Catalysts characterization

The crystal structure were analyzed with an X-ray diffractometer (XRD, D/max-IIIA, Japan) using Cu Ka as the radiation source. The morphologies and microstructure of the prepared samples were observed adopting field emission scanning electron microscope (FE-SEM, MERLIN) and transmission electron microscope (TEM, JEM-2200FS). The UV–visible diffuse reflectance spectra (UV–Vis DRS) were recorded on a UV–Vis spectrometer (U3010, Hitachi) with an integrated sphere attachment. Fluorescence emission spectra were recorded using a fluorescence spectrophotometer (FL-4500, Hitachi) with an excitation wavelength at 350 nm. The Raman spectra were recorded with a LabRAM Aramis confocal laser Micro-Raman spectrometer equipped with an argon ion laser. The surface chemical composition was measured by X-ray photoelectron spectroscopy (XPS, Krato Axis Ultra DLD) with Al Kα X-ray ($h\upsilon$ = 1486.6 eV) at 15 kV and 150 W.

2.3. Photocatalysis process

AO-II was selected as the simulated pollutant to evaluate the photocatalytic activity of $Ag/AgVO_3$. The light visible source was provided by A 300 W Xe arc lamp (PLS-SXE300, Beijing perfectlight Co., Ltd.) equipped with an ultraviolet cutoff filter and the distance between the liquid surface of the suspension and the light source was set about 10 cm. The photodegradation experiments were carried out with the sample powder (100 mg) suspended in AO-II aqueous solution (100 mL, 15 mg L^{-1}) with constant stirring. Prior to the irradiation, the suspensions were magnetically stirred in the dark for 30 min to establish the adsorption/desorption equilibrium. At the given time intervals, about 5 mL of the suspension was taken for further measurement after centrifugation. AO-II photodegradation were analyzed at 484 with UV–vis spectrophotometer (Shimadzu

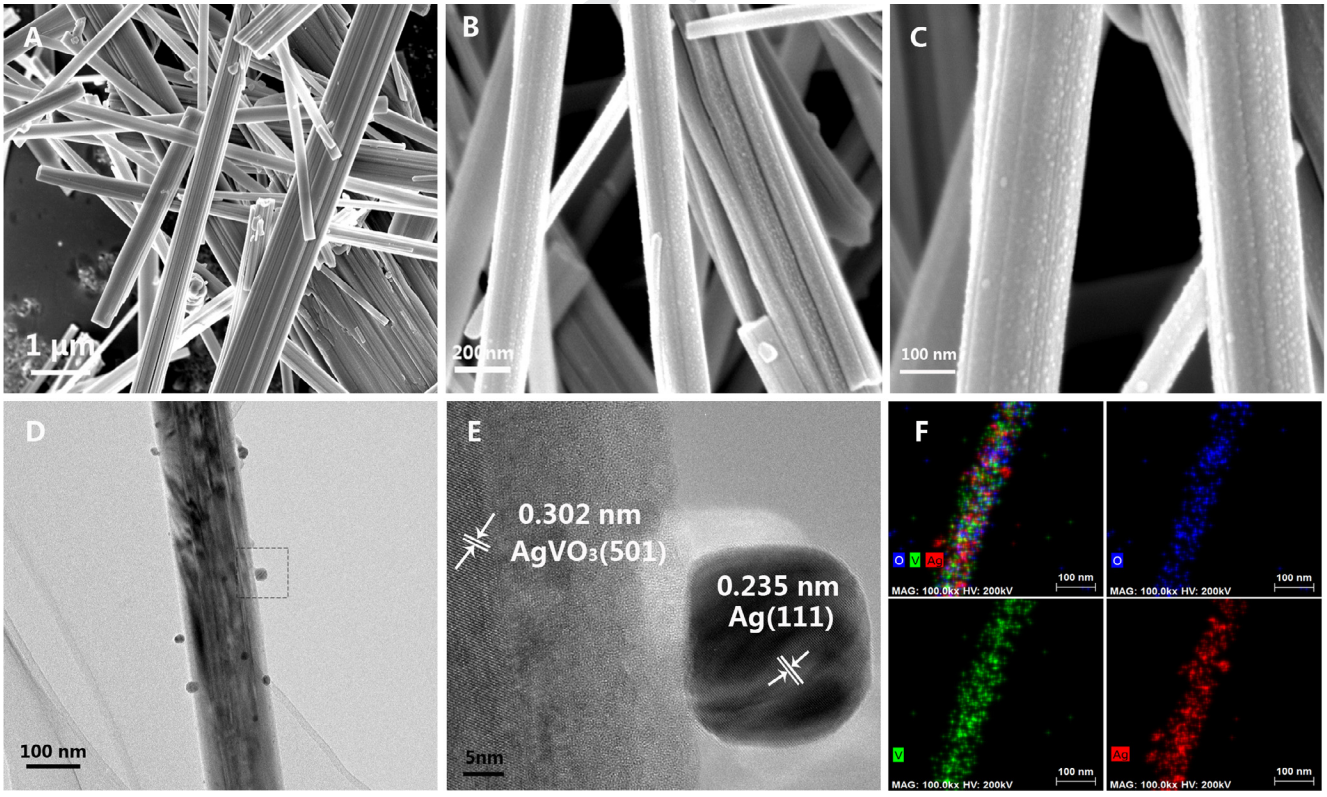

Fig. 1. SEM images of (A) $AgVO_3$; (B) and (C) $Ag/AgVO_3$ with different magnification; (D) TEM images of $Ag/AgVO_3$; (E) HRTEM images of $Ag/AgVO_3$; (F) EDX elemental mapping of the $Ag/AgVO_3$.

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