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Photocatalytic activity of La_2O_3-modified silver vanadates catalyst for Rhodamine B dye degradation under visible light irradiation

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ABSTRACT

La_2O_3/Ag_3VO_4 samples were synthesized by impregnation process and characterized by X-ray diffraction (XRD), scanning electron microscope (SEM), energy-dispersive X-ray spectroscopy (EDS), diffuse reflectance spectroscopy (DRS) and X-ray photoelectron spectroscopy (XPS). The XRD, SEM–EDS and XPS analyses revealed that La^{3+} was dispersed on Ag_3VO_4 in the form of La_2O_3 cluster. The DRS results indicated that the absorption edge of the La^{3+}–Ag_3VO_4 catalyst shifted to longer wavelength. The enhanced photocatalytic activity of La_2O_3/Ag_3VO_4 for Rhodamine B (RhB) dye degradation under visible light irradiation was due to its wider absorption edge and higher separation rate of photo-generated electron and holes. The highest photodegradation efficiency was obtained when the La_2O_3/Ag_3VO_4 catalyst was calcined at 300 °C with 3 wt% La content. The photocatalytic degradation intermediates of the solution were identified by LC/MS.

1. Introduction

Production of clean hydrogen energy and removal of environmental pollutants using photocatalysis technique have received a great deal of interest in recent few decades [1]. Photocatalytic reaction catalyzed by semiconductors has been approved as a promising process for solving energy and environmental issues [2]. Among the semiconductor catalysts, TiO_2 has been studied extensively owing to its special property [3]. However, the wide application of TiO_2 is limited in the condition of solar irradiation due to its wide band gap [4,5]. Therefore, considering energy conservation and environmental pollution issue, it is necessary and indispensable to develop high efficient and visible light-driven photocatalysts. Visible light-induced engineering includes modification of TiO_2 and development of new non-TiO_2-based environmental cleaning materials [6]. Recently, several new environmental cleaning materials including BiOBr [7,8], Bi_2WO_6 [9,10], and $BiVO_4$ [11–13] have been exploited.

Ag_3VO_4 with monoclinic structure has been fabricated and showed photocatalytic activity for water splitting and organic dyes degradation under visible light irradiation [14–16]. However, the photocatalytic activity of Ag_3VO_4 is still low due to its low separation rate of photo-generated electron and holes. Hu and Hu [15] present that the activity of the Ag_3VO_4 is increased by 11 times after doping NiO. It is indicated that the introduction of metal element can enhance the activity of the pure Ag_3VO_4. Recently, it is found that La could be used as cocatalyst, which proved to play a key role in increasing photocatalytic activity [17–20].

In the present work, La_2O_3/Ag_3VO_4 samples with different La concentrations and calcination temperatures are synthesized by impregnation technique. The photocatalytic activities of the samples are evaluated by Rhodamine B (RhB) dye degradation under visible light irradiation. The relationship between the photocatalytic activity and the structure property of the catalysts is discussed. The mechanism of enhanced photocatalytic activities after doping La^{3+} is also given. In addition, the photocatalytic degradation intermediates are identified by LC/MS, and the possible degradation process of RhB dye by La_2O_3/Ag_3VO_4 is proposed.

2. Experimental

2.1. Synthesis of the photocatalysts

Ag_3VO_4 was prepared by precipitation reaction as reported by Hu and Hu [15]. The La_2O_3/Ag_3VO_4 catalysts containing different La contents were prepared by impregnation method with the following procedure: firstly, a stoichiometric amount of La_2O_3 was dissolved in nitric acid (68%) solution. Secondly, 0.5 g of Ag_3VO_4 powder was added into the above $La(NO_3)_3$ solution. Then the suspension was stirred using a glass rod during evaporation of water

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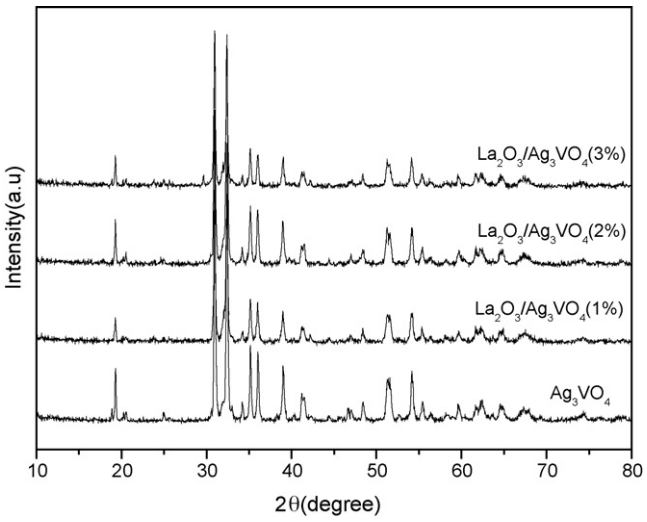

Fig. 1. XRD patterns of La_2O_3/Ag_3VO_4 samples with different La contents.

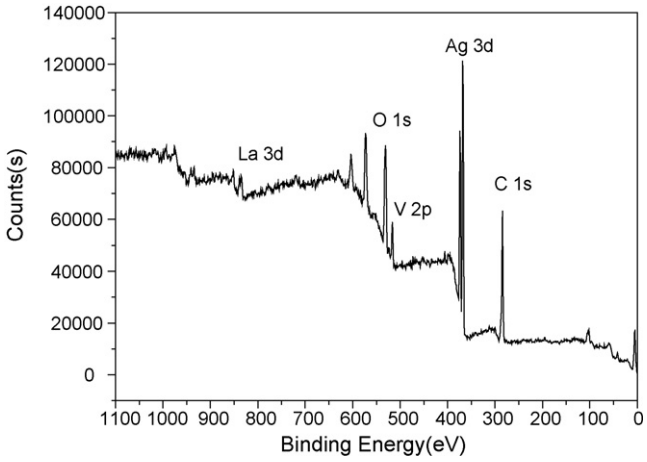

Fig. 3. XPS spectra of La_2O_3/Ag_3VO_4.

in a water bath. The as-synthesized La^{3+}–Ag_3VO_4 was calcined at 300 °C for 4 h. The pure Ag_3VO_4 catalyst was also calcined at 300 °C for 4 h.

2.2. Photocatalysts characterization

The crystalline phases of the prepared catalysts were analyzed by X-ray diffraction (XRD) by Bruker D8 diffractometer with CuKα radiation (λ = 1.54 Å) in the range of $2\theta = 10–80^\circ$.

The surface morphology and particle size of the samples were performed on a field emission scanning electron microscope (FESEM Model JEOL JSM-7001F). The elemental analysis of the photocatalysts was detected by an energy-dispersive X-ray spectrometer (EDS) attached to the SEM.

The X-ray photoelectron spectroscopy (XPS) measurement was performed on the ESCALab MKII spectrometer using MgKα radiation.

The diffuse reflectance spectra (DRS) were performed on a UV-2450 (Shimadzu) instrument in the range of 240–800 nm. $BaSO_4$ was used as the reflectance standard material.

2.3. Photocatalytic activity

The photocatalytic activities of La_2O_3/Ag_3VO_4 catalysts were evaluated by the degradation of RhB dye under visible light irradiation. The photocatalytic reactor consisted of a quartz glass with

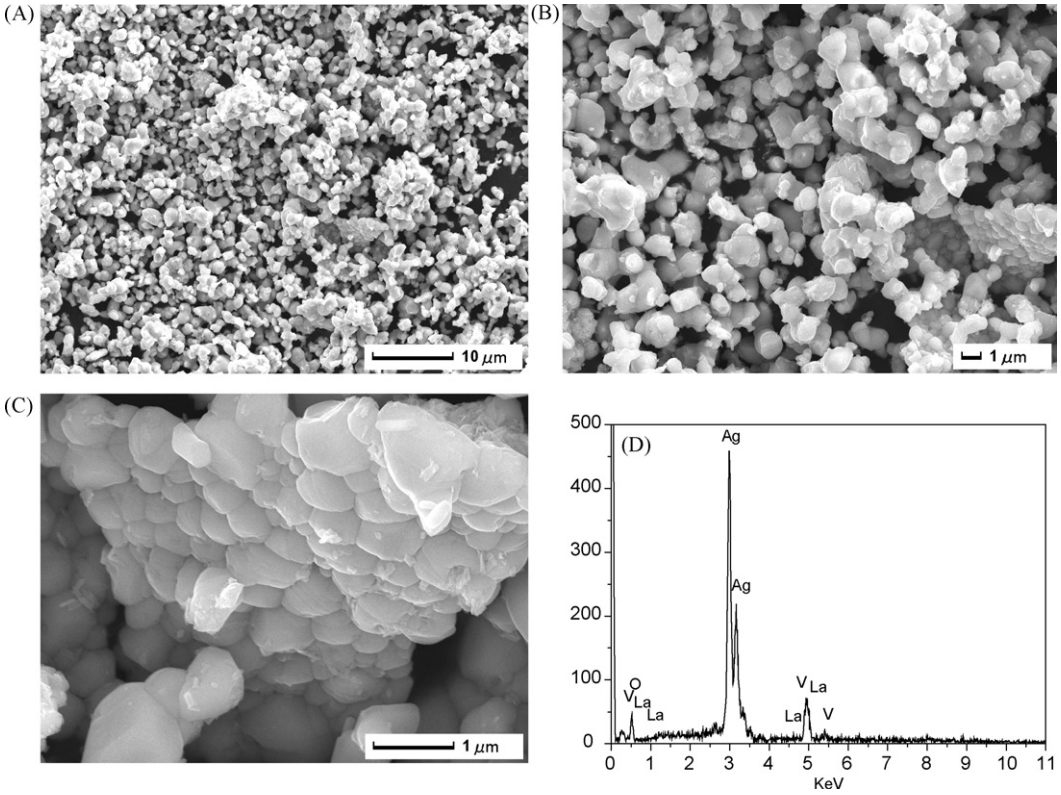

Fig. 2. Images of La_2O_3/Ag_3VO_4 sample (A–C), and (D) EDS spectrum of the La_2O_3/Ag_3VO_4.