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On the electrical conductivity and photocatalytic activity of aluminum-doped zinc oxide

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ABSTRACT

Aluminum-doped zinc oxide (AZO) conductive powders have been successfully prepared by a simple chemical coprecipitation method. The obtained powders with different post-calcining atmospheres were characterized using X-ray powder diffraction (XRD), scanning electron microscopy (SEM/EDS), and dynamic light scattering (DLS). The resistivity-dependent photocatalytic activity was studied by degradation of methyl orange (MO) in aqueous solution. The result showed that photodegradation of methyl orange dyes obeyed the rule of a pseudo first-order kinetics reaction and the AZO photocatalytic activity was related to the resistivity under different post-calcining atmospheres.

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1. Introduction

Industrial wastewater pollution control in the dyeing and printing industries is of importance due to the complex composition, high chromaticity, large emissions, high toxicity, and low biodegradability of wastewater. Weathering of organic dyes through oxidation, hydrolysis or other chemical reactions occurring in the wastewater phase can produce toxic metabolites [1–4]. However, chemical and biological degradation methods have been shown to be ineffective [5]. Currently, oxide semiconductor photocatalysis has attracted a great deal of attention in wastewater treatment because of its high photocatalytic activity, mild reaction conditions, and low energy consumption [6–8].

Among various oxide semiconductor photocatalysts, ZnO has been widely used for its electric and photonic properties and oxidation resistance [9]. Specifically, ZnO nanoparticles can be used for their antibacterial and shielding ultraviolet radiation properties [10,11]. Moreover, ZnO has the photocatalytic capacity to decompose organic dyes to CO_2 and H_2O. We have reported that the ZnO nanoparticles possess antibacterial property and photocatalytic activity [12,13]. However, because ZnO has a wide band gap of about 3.37 eV and a high electron excitation binding energy of about 60 meV at room temperature, ZnO resistivity is very high and can only be excited under UV irradiation, leading to limited application [14], so further research and development is warranted.

The incorporation of other elements into the ZnO photocatalyst could decrease its resistivity and extend its photoresponse, which is found to be an effective route to enhance its photocatalytic performance. Low resistivity and high catalytic efficiency of ZnO can be gained by incorporating the following elements: Co, Sb, Al, In, N and Cu. For example, Chang et al. [15] have reported that the photocatalytic performance of N-doped ZnO was superior to the undoped ZnO under simulated daylight irradiation. Wang et al. [16] have demonstrated that Ag-doped ZnO has improved photocatalytic activities owing to the increased surface defects caused by the enhanced oxygen vacancies. Lim et al. [17] have reported that the Ag/ZnO–SnO_2 catalyst can improve the photodegradation rate of methyl orange. Perazolli et al. [18] have studied the influence of the atmosphere on SnO_2 plus ZnO calcining, verifying that Zn promotes densification due to the formation of oxygen vacancies in the structure. However, to the best of our knowledge, the relationship of electrical conductivity and photocatalytic activity of materials has not been reported so far. So an attempt has been made to study the relationship of electrical conductivity and photocatalytic activity. Additionally, the effect of the post-calcining atmosphere on the conductivity and photocatalytic activity should be investigated.

Herein, we studied the Al-doped ZnO (AZO) photocatalysis due to its widespread attention in recent years not only for its optoelectronic performance, but also for its properties of being nontoxic, pollution-free, thermally stable and an ideal substitute for ITO (indium tin oxide). In this work, we investigated the effect of the different post-calcining

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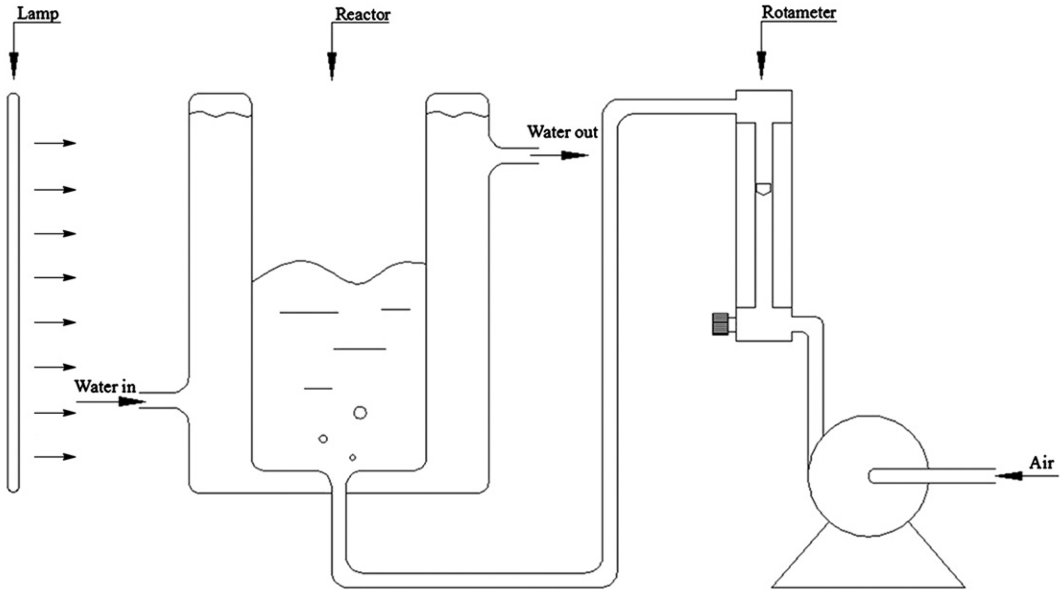

Fig. 1. Apparatus used for the photocatalytic degradation of methyl orange.

atmospheres on AZO electrical conductivity, and also examined the effect of electrical conductivity of AZO on its photocatalytic activity. The photocatalytic activity was studied by degradation of methyl orange (MO) in aqueous solution. The prepared AZO with excellent electrical conductivity showed better photocatalytic activity in degradation of MO dye. Furthermore, the effects of different parameters such as bubbling air and MO initial concentration on MO photodegradation were also explored.

2. Experimental

2.1. Materials

Zinc sulfate ($ZnSO_4 \cdot 7H_2O$), aluminum sulfate ($Al_2(SO_4)_3 \cdot 18H_2O$), sodium hydroxide (NaOH), anhydrous sodium carbonate (Na_2CO_3) and polyethylene glycol 400 (PEG-400) were all of analytical grade and purchased from Wuxi Chemical Co., Ltd. Methyl orange (MO) was used to measure the AZO photocatalytic activity. Deionized water was used throughout the experiments.

2.2. Preparation of AZO conductive powders

2.2.1. Precursor preparation

AZO precursors were prepared by a simple chemical coprecipitation route. Generally, $ZnSO_4 \cdot 7H_2O$ and $Al_2(SO_4)_3 \cdot 18H_2O$ with the molar ratio of n (Al)/n (ZnO) = 1.5 at.% were firstly dissolved in deionized water, then poured into a three-necked flask, PEG-400 was added at a mass ratio to $ZnSO_4 \cdot 7H_2O$ mass ratio of 1.5 wt.%. A mixed alkali solution of NaOH and Na_2CO_3 with the concentration ratio of $c(NaOH)/c(Na_2CO_3) = 2$ was added to the three-necked flask drop by drop at 60 °C with stirring (400 rpm) until pH reached a value of 7.2 (measured by a digital pH meter). After a 150 min precipitation, the precipitate was filtered and washed several times with distilled water, and followed by drying a few hours in an electric oven at about 100 °C to obtain the dried powders. The dried powders were pre-calcinated at 400 °C for 2 h in order to make $ZnCO_3 \cdot 2Zn(OH)_2$ and $Al(OH)_3$ decompose into oxides and then ground in a mortar for about 2 min.

2.2.2. Post-calcination process for precursor

Sample A: These obtained powders were post-calcinated at 900 °C in normal air for 2 h with a constant heating rate of 10 °C/min.

Sample B: Post-calcinated at 900 °C for 2 h in an inert argon (Ar) atmosphere with a constant heating rate of 10 °C/min.

Sample C: Post-calcinated at 900 °C for 2 h in reducing atmosphere (CO) with a constant heating rate of 10 °C/min.

2.2.3. Characterization

The structure of the AZO conductive powders was investigated by X-ray diffraction (XRD) on the Bruker D8ADVANCE X-ray diffractometer at a voltage of 40 kV with Cu Kα radiation ($\lambda = 1.5406$ Å) in the 2θ ranging from 27° to 80° and the step width in terms of Delta_2_theta was set to 0.01°. The microscopy and the composition of the products were examined by the scanning electron microscopy (SEM, S-4700 field emission, Hitachi, Ltd) and an accessory (EDS) of SEM. The particle size distribution of the AZO conductive powder was measured using Malvern HPPS5001 laser particle-size analyzer (DLS) at 25 °C. Before measuring DLS, the AZO conductive powders were dispersed in ethanol under ultrasonic condition (800 W) for 30 min. The obtained suspension was diluted to a nearly colorless and transparent liquid to measure. The resistivity was tested on a RTS-9 four-point probe resistivity

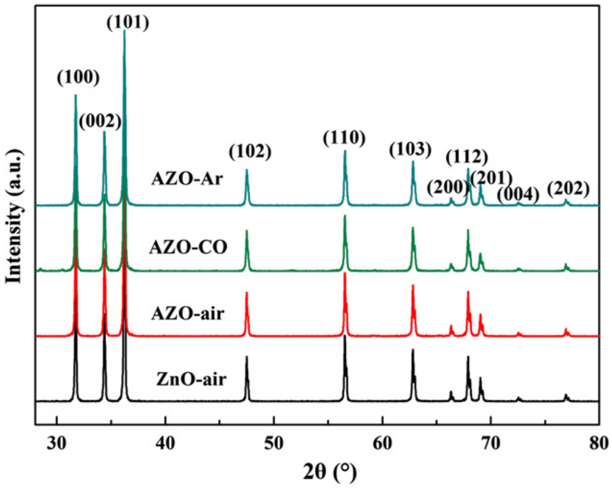

Fig. 2. XRD patterns of ZnO powder and AZO samples with post-calcining under different atmospheres.

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