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Synthesis, characterization, and photocatalytic activities of titanate nanotubes surface-decorated by zinc oxide nanoparticles

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ABSTRACT

Nanoscaled zinc oxide (ZnO) particles with different amounts are coated on titanate nanotubes (TNTs) by a facile chemical method at room temperature. The characterizations of XPS, TEM, XRD and UV–vis spectra confirm that pure hexagonal wurtzite ZnO nanoparticles with an average size of about 9 nm are distributed on the surfaces of TNTs evenly and attached strongly. The photocatalytic activities of the ZnO–TNTs nanocomposite are superior to those of P25, ZnO, TNTs and ZnO–anatase TiO_2 (TNP) nanocomposite in the oxidation of rhodamine B under UV light irradiation. A comparison of the photocatalytic activities between different catalysts is discussed. Furthermore, we also find that the ZnO–TNT nanocomposite shows very favorable recycle use potential, because they have a high sedimentation rate and their photocatalytic activity is only slightly decreased even after five times of repeated uses.

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1. Introduction

Recently, titanates with one-dimensional (1D) TiO_2 nanostructures such as nanotubes, nanowires and nanofibers [1–4] have attracted a lot of attention due to their significant potential applications. Among 1D titanate nanostructures, titanate nanotubes (TNTs), which are of layer structure with a hollow cavity, possess unique physicochemical properties and have become one of the most promising materials in various fields [5,6]. A great development has been achieved in the study of synthesis and the structure of these nanotubes [7–10]. In the past few years, increasing interests have being focused on the TNTs decorated with active catalysts, including metal ions and semiconductors in the application of catalysis process, such as photocatalysis, electrocatalysis and photoelectrocatalysis [11–13]. In fact, the high cation-exchange character as well as the particular tubular structure makes TNTs very suitable to act as the substrate and carrier for different catalysts that need to be immobilized. Furthermore, the decoration of enzymes [14], nanoparticles [15,16] and metal ion [17] with TNTs has resulted in better catalysis activities, because the unique physical properties of TNTs, such as open mesoporous morphology and high specific surface area, make the reagents easier to transport during the catalytic reaction [5]. All these interesting properties of TNTs encourage us to investigate it as a support for different catalysts.

Recently, ZnO becomes one of the most widely studied multifunctional nanocrystalline semiconductors and attracts attention for its wide range of applications, such as solar cells, luminescent, electrical and chemical sensors due to the wide band gap (3.2 eV) and large exciton binding energy (60 meV) [18–21]. The ZnO–TiO_2 nanoparticles have been synthesized successfully and the researches have proved that utilization of the nanocrystalline TiO_2 coupled with nanosized ZnO could improve their photocatalytic efficiency due to the synergistic effect on photocatalytic properties [22–25].

TNTs have several distinct advantages to be the substrate of ZnO–TNTs nanocomposite. Firstly, the high cation exchange properties make Zn^{2+} ions adsorb on TNTs more strongly and evenly [26]. Secondly, the surface area of TNTs, which include the inner-cavity and outer surface, is large enough to accommodate more ZnO nanoparticles [27]. Thirdly, TNTs are more convenient than spherical powder catalysts, such as P25, for separating catalysts from suspended solution because of the high sedimentation rate [28]. Some studies have shown the use of CdS–TNTs [29,30] or SnO_2–TNTs [31] nanocomposite can enhance the performance in photocatalysis. However, to the best of our knowledge, there are few reports regarding the properties of TNTs coupled with ZnO nanoparticles and their repeated use in photocatalysis. Herein, we report the preparation of ZnO–TNTs nanocomposite by a facile chemical method at room temperature. Their characterizations of XPS, TEM, XRD and UV–vis spectra are also discussed. The prepared nanocomposite show excellent photocatalytic properties on the degradation of rhodamine B (RhB) under UV irradiation. Further-

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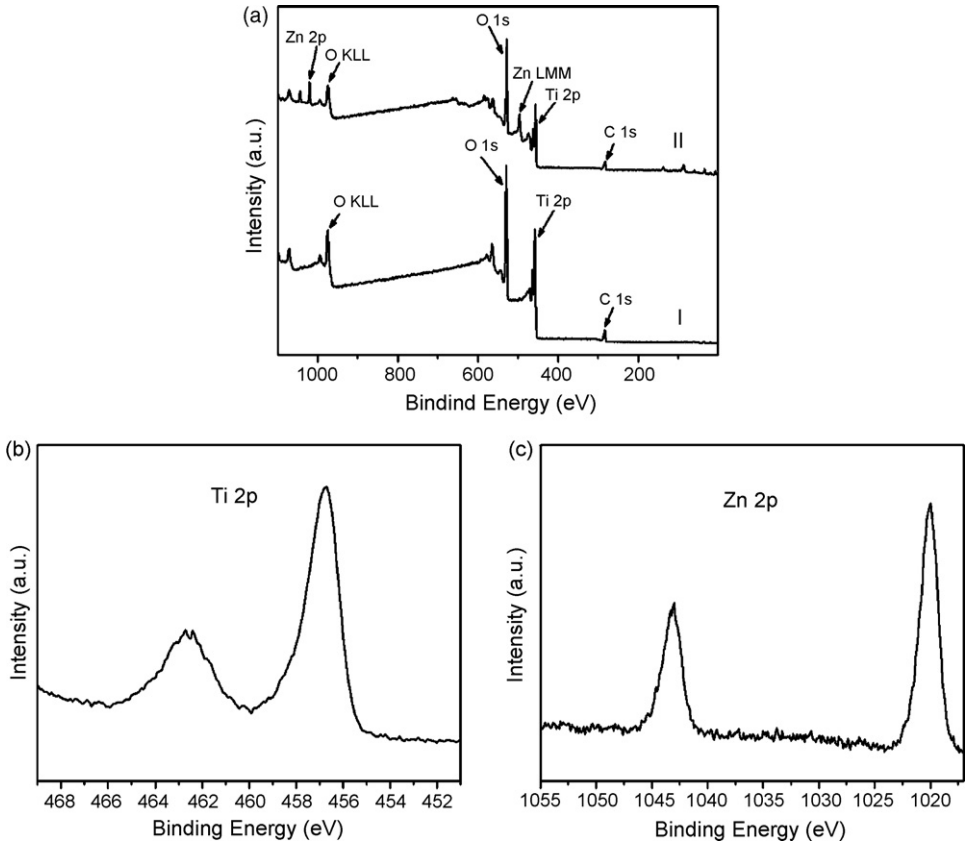

Fig. 1. (a) XPS survey spectra of TNTs (I) and ZnO–TNTs (II), (b) Ti 2p XPS spectra of ZnO(20 wt.%)–TNTs, (c) Zn 2p XPS spectra of ZnO(20 wt.%)–TNTs.

more, the recycle of ZnO–TNTs is more convenient and no obvious decrease in the photocatalytic activity is observed for the recycled ZnO–TNTs.

2. Experimental

2.1. Reagents and measurement

Zinc acetate, sodium hydroxide and RhB were of analytical reagent grade purchased from Guangzhou Chemical Reagents Factory (Guangzhou, China) and used without further purification. Deionized water was used in all aqueous solution preparations and washings.

2.2. Synthesis

2.2.1. Titanate nanotubes synthesis

TNTs were prepared following the literature procedure [32]. Briefly, 2 g of commercial anatase TiO_2 nanoparticles (TNP) was added to 50 mL of 10 M NaOH solution and heated for 24 h at 130 °C in a Teflon-lined autoclave. After cooled naturally in air, the mixture was centrifuged at a speed of 4000 rpm and the precipitates were collected. The white powder was thoroughly washed with water then with 0.1 M HCl, followed by vacuum drying at 70 °C.

2.2.2. Preparation of ZnO–TNTs nanocomposite

The ZnO–TNTs nanocomposite was synthesized by the following method. Stoichiometric amount of pure TNTs and zinc acetate was dispersed in 50 mL absolute ethanol by stirring for 6 h. Then the absolute ethanol solution of sodium hydroxide (50 mL, 0.2 M) was gradually added with vigorous stirring at room temperature. After the mixture was stirred for 10 h, a white precipitate was obtained. Then the white powder was collected by centrifugation and then washed several times with deionized water and ethanol for several times. In addition, the ZnO–TNP was prepared by using in a similar way in order to make a comparison of the photocatalytic properties with that of the ZnO–TNTs.

2.3. Characterizations

X-ray photoelectron spectroscopy (XPS) measurements were done with AXis Ultra^{DLD} (Kratos). The powder X-ray diffraction (XRD) patterns were recorded using an XD-3A Cu Kα X-ray diffrac-

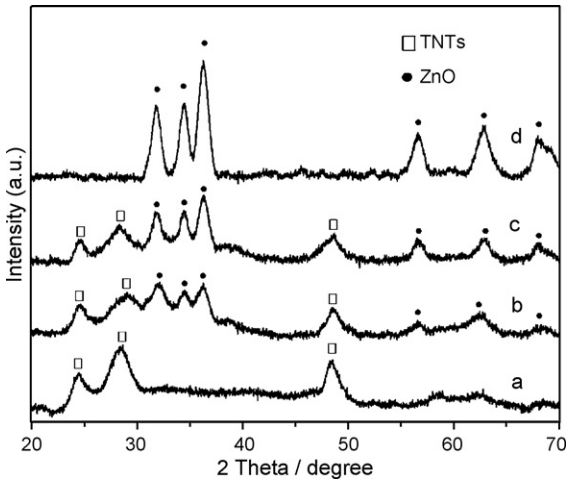

Fig. 2. X-ray diffraction patterns of (a) TNTs, (b) ZnO(10 wt.%)–TNTs, (c) ZnO(20 wt.%)–TNTs and (d) ZnO nanoparticles.

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