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Sol–gel synthesis of novel cobalt doped zinc tin oxide composite for photocatalytic application

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Abstract

A novel photocatalyst based on cobalt doped zinc tin oxide is proposed. Cobalt doped zinc tin oxide thin films were deposited using a sol–gel deposition method and characterized by scanning electron microscopy (SEM), X-ray diffraction, Raman spectroscopy, photoluminescence emission measurement and UV–vis spectroscopy. It was found that the addition of Co into the zinc tin oxide does influence the structural and optical properties of the thin films and increases the overall photocatalytic degradation of methylene blue.

Keywords: Cobalt; Zinc tin oxide; Photocatalyst; Methylene blue

1. Introduction

Recently, there have been great interest in metal oxides due to its potential application in a wide range of fields [1,2]. For example, zinc oxide has been used in photocatalytic applications [3,4] and tin oxide has been extensively employed as transparent conductive oxides in dye sensitized solar cells [5,6]. The combination of Zn and Sn has been shown to produce zinc tin oxide (ZTO) that is an n-type semiconducting material with a wide band gap energy of $\sim$3.6 eV and high electron mobility [7], which makes it suitable for photovoltaic devices, sensors, surface functionalization, and photocatalytic applications [8].

In the past, ZTO thin films have been deposited by solid phase decomposition of Zn and SnO powders in a furnace [9,10]. Although high quality ZTO thin films can be obtained by solid phase crystallization process, it requires deposition temperature far greater than the melting temperature of most glass and plastic substrates and therefore limits the substrate selection. ZTO thin films can also be deposited by sputter coater by using pre-sintered ZTO target that can be scaled-up for large area deposition. However, the sputtering process requires expensive sputter targets and involves complicated vacuum set-up [11,12]. Alternatively, ZTO coatings have also been deposited by solution-based method such as sol–gel deposition [13] and hydrothermal growth within teflon reactors [14]. Solution-based deposition method offers low-cost, large-area deposition with possible integration into inkjet printing deposition technologies for roll-to-roll processes.

Previous studies have revealed that the structural and optoelectronics properties of ZTO thin films are highly influenced by deposition conditions. For example, structure properties of ZTO can be modified by addition of rare earth materials such as dysprosium without significant effect on its band-to-band emission [15], whereas the introduction of europium reduce the strains within the lattice [16]. In the past, cobalt has been incorporated into individual ZnO and SnO to produce diluted magnetic [17] and ferromagnetic semiconductors [18], respectively. To the best of our knowledge, there has been little or no study on Co doped ZTO.

In the present study, a new type of photocatalyst, Co doped ZTO thin films were synthesized by sol–gel deposition method. The investigated thin films were extensively characterized using scanning electron microscope, energy dispersive spectroscopy, Raman spectroscopy, UV–vis spectroscopy and photoluminescence measurements.

2. Experimental

All chemicals used during this study were purchased from Sigma Aldrich and used without further purification. First, zinc

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acetate dehydrate and tin (II) chloride were dissolved in isopropanol (IPA), with an equimolar ratio of Zn:Sn was maintained constant throughout this work. The precursor solution was magnetically mixed at 60 °C for 2 h. To this solution, 1–5 at% of cobalt acetate was added and further mixed for 3 h. The resultant solutions were left to age for at least 2 days before deposition.

Acetone and IPA were used to clean the corning glasses (Eagle 2000) with the aid of ultrasonic agitation. The prepared sol precursors were spun onto the glass substrates using a spin coater, operating at spin speed of 3000 rpm. Then the deposited precursor undergoes two sintering process at 250 °C, to evaporate the solvents and at 550 °C to crystallize the films. Each spin and sintering cycle yielded 60 nm of ZTO:Co film and was repeated five times to yield a thickness of around 300 nm.

The morphology and composition of the synthesized ZTO:Co thin films were evaluated using a FEI Quanta 400F environmental scanning microscope, equipped with an energy dispersive spectroscopy (EDS). A Siemens D5000 X-ray diffractometer with Cu Kα radiation was used to determine the crystalline orientation and grain size were determined from.

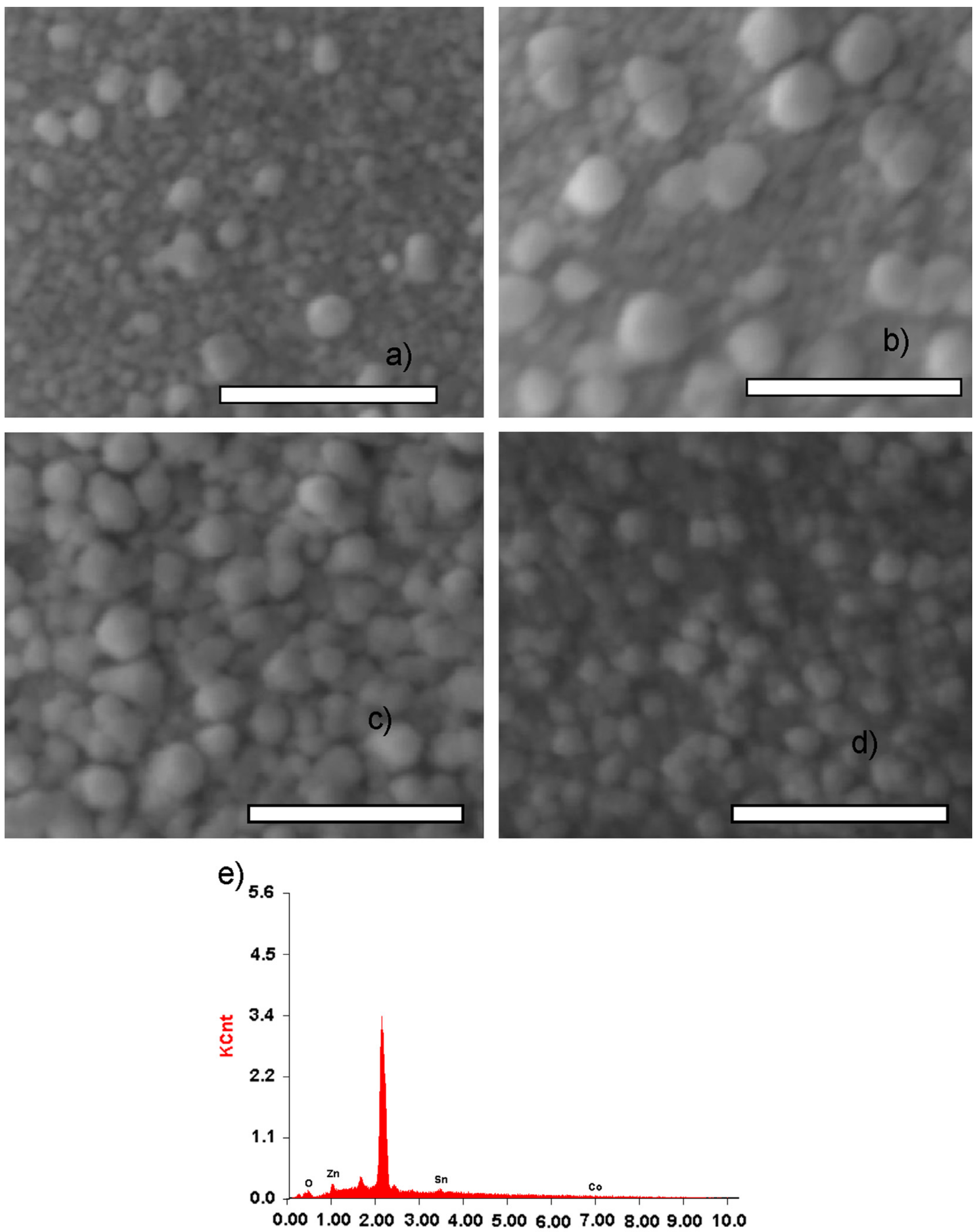

Fig. 1. Scanning electron microscopy images of ZTO:Co thin films doped at Co concentration of (a) 1 at%, (b) 2 at%, (c) 3 at%, and (d) 5 at% (Scale bar 1 μm) and (e) EDS composition analysis.