



Enhanced visible-light photocatalytic activity of strontium-doped zinc oxide nanoparticles

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ABSTRACT

Strontium-doped zinc oxide nanoparticles ($Zn_{1-x}Sr_xO$ NPs; $x=0, 0.02, 0.04, \text{ and } 0.06$) were synthesized by a sol-gel method. Transmission electron microscopy (TEM) and scanning electron microscopy (SEM) images showed NPs with nearly spherical shapes, with sizes from 27 to 41 nm for high Sr concentration and undoped ZnO NPs, respectively. X-ray diffraction (XRD) patterns, selected area electron diffraction (SAED) patterns, and Raman spectra indicated that the undoped and Sr-doped ZnO NPs were crystallized in a hexagonal wurtzite structure. However, the Raman results revealed a decrease in the crystalline quality with an increase in the Sr concentration in the ZnO structure. Evidence of dopant incorporation is demonstrated by X-ray photoelectron spectroscopy (XPS) of the Sr-doped ZnO NPs. From the results of optical characterizations, the band-gap values of the $Zn_{0.98}Sr_{0.02}O$ and $Zn_{0.96}Sr_{0.04}O$ NPs decreased, while the band-gap value of the $Zn_{0.94}Sr_{0.06}O$ NPs increased in comparison to the band-gap value of the undoped ZnO NPs. Finally, the obtained NPs were used as a photocatalyst to remove methylene blue (MB). Observations showed that the efficiency of the photocatalyst activity of the ZnO NPs was significantly increased by increasing the Sr, but until an optimum concentration.

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1. Introduction

Today, environmental problems are very important issues for the governments in the development countries.

One of these problems is water pollution. Currently, organic dyes and their effluents have become some of the main sources of water pollution; these organic dyes escape from traditional wastewater treatment plants and remain in the water because of their high stability against light, temperature, chemicals, and microbial attack. Photocatalytic technique is a cheap and effective technique to solve this problem. Significant advances have recently been made in the area of semiconductor nanostructures

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for photocatalytic applications [1–4]. A photocatalyst is also called a photochemical catalyst, and its function is similar to that of chlorophyll in photosynthesis. In a photocatalytic system, a photo-induced molecular transformation or reaction takes place at the surface of the catalyst. A basic mechanism of a photocatalytic reaction is the generation of an electron–hole pair, which can be described as follows. When a photocatalyst is illuminated by a light stronger than its band gap energy, electron–hole pairs diffuse out to the surface of the photocatalyst and participate in a chemical reaction with an electron donor and acceptor.

A high redox potential, a direct band-gap of 3.37 eV at room temperature, a high exciton binding energy of 60 meV, superior physical and chemical stability, inexpensiveness, easy synthesis, and non-toxicity were identified as the main reasons for the wide acceptability of ZnO materials compared to other semiconductor photocatalysts. Therefore, ZnO can be used as a photocatalyst material to remove organic dyes from wastewater with high efficiency [5–10]. In order to utilize solar radiation more effectively, the development of a wide band gap ZnO is necessary. In order to obtain different properties and applications for ZnO, researchers carried out doping in ZnO. Alkaline earth metal elements can be taken as the candidate dopants to regulating and controlling the photocatalytic properties. In fact, choosing a cation as doping material with bigger ion radius than Zn^{2+} radius results in the creation of significant lattice defects because of the charge compensation and the ionic radius mismatch between the cation and Zn^{2+} , which could affect photocatalytic properties of ZnO. We believed that Sr^{2+} with so bigger radius ($R_{\text{Sr}^{2+}} = 2.45 \text{ \AA}$) than Zn^{2+} radius ($R_{\text{Zn}^{2+}} = 0.74 \text{ \AA}$) can affect optical properties of ZnO. It is well known that optical and photocatalytic properties have a direct relationship. Therefore, we expect to observe these effects on degradation efficiency of dyes by Sr-doped ZnO nanostructures.

In this study, a simple sol–gel method was used to synthesize ZnO NPs with a narrow size distribution that was doped by Sr with different concentrations. Then, effects of Sr on crystallinity and optical properties were investigated. Finally, the obtained products were used as photocatalyst materials under visible-light irradiation to remove methylene blue (MB), which is one of the dye materials.

2. Experimental

To begin the synthesis of Sr-doped ZnO-NPs, analytical grade zinc nitrate hexahydrate $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, strontium nitrate $\text{Sr}(\text{NO}_3)_2$, gelatin (type B from bovine skin), and distilled water were used as starting materials. All of the materials used were purchased from Sigma-Aldrich. The precursors were taken in the stoichiometric amounts of $\text{Zn}_{1-x}\text{Sr}_x\text{O}$ ($x=0, 0.02, 0.04, \text{ and } 0.06$) to obtain final products. First, a gelatin solution was prepared by adding gelatin (3.65 g) to distilled water at $60 \text{ }^\circ\text{C}$. The metal nitrates were dissolved separately in a minimal amount of distilled water at room temperature, and then these were added to the gelatin solution. After that, the compound solutions

were stirred and heated at $80 \text{ }^\circ\text{C}$ until gels were obtained. The gels were calcined at $550 \text{ }^\circ\text{C}$ for 5 h, at a heating rate of $2 \text{ }^\circ\text{C}/\text{min}$. The resulting powders were characterized using several tools to check their quality.

X-ray diffraction (XRD) (Philips, X'Pert, $\text{CuK}\alpha$) was used to evaluate the phase characteristics of the samples. Transmission electron microscopy (TEM, Hitachi H-7100) and selected areas electron diffraction (SAED) were employed to characterize morphology and structural properties of the NPs. Elemental analyses of the products were conducted using an energy-dispersive X-ray spectroscope (EDX), which was attached to a field emission scanning electron microscope (FESEM, Quanta 200 F), and using X-ray photoelectron spectroscopy (XPS; VG-Microtech ESCA-2000). The XPS spectra were recorded using $\text{Mg-K}\alpha$ radiation (1256.6 eV). Raman measurements (Jobin Yvon Horiba HR 800 UV) were carried out to study the crystallinity of the obtained products. An Ar ion laser with an emission wavelength of 514.5 nm was used for the Raman measurements. The optical properties of the ZnO-NPs were characterized at room temperature using a UV–vis (PerkinElmer, Inc.) and photoluminescence (PL, Jobin Yvon Horiba HR 800 UV) spectrometers using a He–Cd laser with a wavelength of 325 nm.

The photocatalytic performance of the as-prepared samples was evaluated using the photocatalytic degradation of MB under visible-light irradiation. Here, 10 mg of the obtained material was dispersed in 30 ml of the MB aqueous solution (10 mg/l). The mixed suspension was magnetically stirred for 30 min in dark to reach an adsorption–desorption equilibrium. Under ambient conditions and stirring, the mixed suspension was exposed to visible-light irradiation produced by a Xenon lamp (500 W) used as the visible-light source for different times (0–120 min). At certain time intervals, 2.5 ml of the mixed suspension was extracted and centrifugated to remove the photocatalyst. The degradation process was monitored by measuring the absorption of MB in the filtrate at 664 nm using a UV–vis absorption spectrometer.

3. Results and discussion

The XRD patterns of the NPs in the range of $2\theta = 30\text{--}70^\circ$ are shown in Fig. 1. All of the detectable peaks could be indexed as the ZnO wurtzite structure (PDF card no.: 00-005-0664). No other peaks such as Sr, SrO, and ZnSrO were detected. Fig. 1(b) shows a shift in the (101) peaks for the samples that were doped with Sr elements in the ZnO matrix, compared to the undoped ZnO-NPs. The ionic radius of the substitute Sr^{2+} ($R_{\text{Sr}^{2+}} = 2.45 \text{ \AA}$) is bigger than that of Zn^{2+} ($R_{\text{Zn}^{2+}} = 0.74 \text{ \AA}$). Thus, doping with Sr caused a slight shift in the (101) XRD peaks toward lower diffraction angles. The extent of these shifts was dependent on the Sr content in the NPs. This result provides indirect evidence that strontium is incorporated into the crystal structure, causing the ZnO crystal lattice to expand.

The crystallite sizes of the ZnO-NPs were determined by means of an X-ray line-broadening method, using the Scherrer equation

$$D = (k\lambda/\beta_{hkl} \cos \theta) \quad (1)$$

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