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Synthesis and characterization of In doped ZnO thin film as efficient transparent conducting oxide candidate

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ABSTRACT

In this work, we have successfully prepared the highly conducting and transparent In doped ZnO thin films on glass substrate using sol-gel spin-coating technique. Indium was incorporated with different concentrations of 1, 2, and 4 at.%. The effect of indium doping on the structural, optical and electrical properties of the produced films have been investigated. X-ray diffraction analysis showed that all the films were polycrystalline with a hexagonal würtzite structure.The growth along (002) orientation was only preferred for 2 at.% doping concentration. The transparency of In doped ZnO thin films varied from 70 to 92% in visible range. Zinc oxide thin film doped with 4 at.% concentration revealed the largest grain size, the lowest optical gap, the highest intrinsic defects amount, and the lowest resistivity which was found to be 6.10×10^{-4} Ω cm. These In doped ZnO thin films can have big interest in solar cell industry.

1. Introduction

Zinc oxide is a semiconductor material that has occupied a great interest in many researches. This promising material is highly transparent in visible region with a wide band gap of 3.37 eV and excitonic energy of 60 meV at room temperature. Moreover, zinc oxide has good stability and adherates well on different substrates. This oxide serves to develop many industrial products such as solar cells, piezoelectric devices, gas sensors, light emitting diodes (LEDs), and heat mirrors [1–6].

Sol-gel spin-coating process is one of the common techniques used to synthesize zinc oxide thin films. This simple and low cost technique allows controling many parameters such as doping concentration, thickness, and samples homogeneity [7–9]. Doping permits to improve zinc oxide properties especially transparency and conductivity. Ionic radius and electronegativity of dopant have an interesting effect on the desired properties [4].

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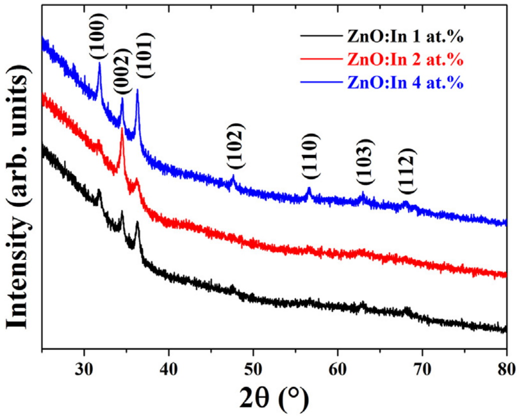

Fig. 1. XRD patterns of In doped zinc oxide thin films.

Table 1
The diffraction angle 2θ, FWHM, and the grain size for In doped ZnO thin films in the three orientations (100), (002), and (101).

Samples	2θ (°)			FWHM (°)			Grain size (nm)		
	(100)	(002)	(101)	(100)	(002)	(101)	(100)	(002)	(101)
In 1 at.%	31.82	34.47	36.33	0.4093	0.2558	0.2047	20.18	32.51	40.84
In 2 at.%	31.77	34.47	36.19	0.6140	0.3070	0.4093	13.45	27.09	20.42
In 4 at.%	31.84	34.50	36.30	0.2558	0.1023	0.2047	32.29	81.31	40.84

In this work, we aim to obtain In doped ZnO thin films on glass substrates with electrical resistivity of the order of $10^{-4}\,\Omega$ cm and high transparency (≈90%) using sol-gel spin-coating technique. We expose the effect of indium doping concentration on the structural, optical, and electrical properties of In doped ZnO thin films.

2. Experimental

Sol-gel spin-coating method was utilized to elaborate indium doped zinc oxide thin films. Indium chloride ($InCl_3$) was introduced with different concentrations of 1 at.%, 2 at.%, and 4 at.%. Zinc acetate dihydrate ($Zn(CH_3COO)_2 \cdot 2H_2O$) was dissolved in 5 ml of isopropanol ($(CH_3)_2CHOH$) and the indium chloride was dissolved in 5 ml of ethanol (CH_3CH_2OH), separately. The two solutions of zinc acetate and indium chloride were mixed together in order to obtain a final solution of 10 ml with a molarity of 0.2 M for each indium doping concentration. Few drops of mono-ethanolamine (MEA) were added to stabilize the obtained solutions. Simultaneous stirring and heating of each solution at 60 °C for 2 h using a hot plate stirrer allowed reaching a transparent and homogenous solution.

A small volume of each solution was deposited and rotated on glass substrate for 5 s with a velocity of 2000 rpm producing a thin film which was heated for ten minutes at 300 °C in order to remove organic residuals such as isopropanol, ethanol and MEA. The repetition of the previous operation five times permitted attaining the desired thickness. Good crystallization of all thin films required annealing at 500 °C during 1 h.

Thin films structural properties were analyzed in the θ–2θ configuration using an Xpert powder diffractometer with Cu X-ray radiation (λ = 1.5406 Å). UV–vis SHIMADZU UV-3101PC spectrophotometer and Perkin Elmer LS 55 luminescence spectrometer were used to determine the optical transmittance and photoluminescence emission at room temperature, respectively. Samples electrical resistivity and thickness measurements were based on JANDEL four-point probe and Dektak 150 profilometer, respectively.

3. Results and discussion

3.1. Structural properties

X-ray diffraction analysis (Fig. 1) showed that indium doped ZnO thin films were polycrystalline with hexagonal würtzite structure. No diffraction peaks corresponding secondary phases such as In_2O_3 or $Zn_2In_2O_3$ were observed as a result of indium incorporation [10,11]. These thin films were preferentially grown along the (101) plane except for In 2 at.% doping concentration where the growth was more enhanced along the (002) plane and indium increasing content improved the crystallinity. Diffraction angle position of peaks corresponding the (002) plane in all samples was shifted towards higher value with respect to standard value 34.45° displayed in JPCDS Card No. 36-1451 (see Table 1) which confirmed the existence of stress in these thin films.