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Synthesis and characterization of pure ZnO and La-doped ZnO ($Zn_{0.98}La_{0.02}O$) films via novel sol-gel screen- printing method

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ABSTRACT

In present work ZnO and $Zn_{0.98}La_{0.02}O$ films were synthesized by sol-gel screen –printing method on glass substrate. The prepared films were investigated by XRD, SEM, EDAX, UV–vis and two probe methods for structural, optical and electrical properties. The X-ray diffraction results showed that the films were polycrystalline with hexagonal wurtzite structure and had preferred growth of grains along the (100) crystallographic direction. SEM images of pure ZnO and doped $Zn_{0.98}La_{0.02}O$ films indicate the porous and unsymmetrical distribution of grains. The EDAX spectra showed the successful incorporation of La ions with ZnO lattice. The band gap of the films was determined through UV–vis Spectroscopy. It is observed that band gap increases with La doping. The electrical conductivity measurement was carried out via two probe method which reveals the semiconducting behavior of the films.

1. Introduction

ZnO a II-VI semiconductor compound is extensively studied topic of research because of its wide energy band gap of 3.2–3.37 eV at room temperature and large excitation binding energy of 60 MeV. Several researches have been done on this material till date. Every year research is found on different journals on it. ZnO is Low cost and non-toxic in nature so environmental friendly as compared to other metal oxides [1]. It has wide applications in various fields such as optoelectronics, antibacterial, gas sensing and cancer treatment [2,3]. Band gap of ZnO material can be adjusted by appropriate doping material (e.g. Ni, Mn, Al, Cu, Fe, Ce, Cd and La etc.) [4]. Doping of Rare earth elements in ZnO is a research focus in recent years. Doping of rare earth elements via sol-gel screen printing is quite rare. Usually methods that are used to prepare thin films by sol-gel are spin coating and dip coating. To date ZnO thin films have been prepared by variety of techniques such as RF magnetron sputtering [5], DC magnetron sputtering [6], pulsed laser deposition [7], metal organic CVD (MOCVD) [8], chemical bath deposition (CBD) [9], molecular beam epitaxy [10], spray pyrolysis [11], chemical vapor deposition [12,13], electrochemical deposition [14], sol-gel [15], screen printing [16].

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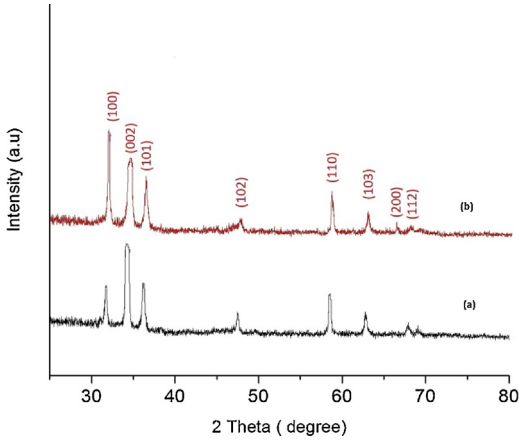

Fig. 1. X- ray diffraction pattern of (a) Pure ZnO film and (b) La-doped ZnO ($Zn_{0.98}La_{0.02}O$) film.

In the present study for the first time we report the preparation of ZnO and La-doped ZnO ($Zn_{0.98}La_{0.02}O$) films via sol-gel screen printing method. It is particularly efficient in producing films on various substrates at economic cost and easy to carry out in laboratories.

In the present report ZnO and La-doped ZnO ($Zn_{0.98}La_{0.02}O$) films were prepared by using novel, simple and economic sol-gel screen printing process and their physical properties were carried out via XRD, SEM, EDAX, UV–vis Spectroscopy and electrical conductivity measurement employing two probe technique.

2. Experimental procedure

In this work pure ZnO and doped ZnO ($Zn_{0.98}La_{0.02}O$) films were prepared via sol-gel screen printing technique. All chemicals used here were AR grade with 99.99% purity. For the preparation of pure ZnO the materials that were used zinc acetate dihydrate Zn $(CH_3COO)_2 \cdot 2H_2O$, monoethanolamine (MEA: C_2H_7NO) as a stabilizer and ethylene glycol as a binder. Stoichiometric amount of zinc acetate dihydrate was dissolved in 20 ml of ethylene glycol with 10 ml of MEA in a stirrer and it formed transparent sol. Sol was converted to a clear gel by continuous stirring while heating at 100 °C for several hours. The pH of the solution was adjusted to value between 7–8 by adding ammonia solution. The precursor for the preparation of La-doped ZnO films were zinc acetate dihydrate, lanthanum trinitrate hexahydrate [La $(NO_3)_3.6H_2O$] MEA and ethylene glycol as binder. The mixture of zinc Acetate (98%) and Lanthanum trinitrate (2%) was dissolved in 20 ml of ethylene glycol with 10 ml of MEA and same process was followed for preparation of La-doped ZnO films as used for preparation of pure ZnO films. pH was maintained between 7–8. The prepared gel of pure and doped ZnO were kept for 72 h at room temperature then films were deposited evenly on pre-cleaned glass substrates using screen printing method. After deposition, these films were heated for 1 h at 100 °C and then sintered at 500 °C for 10 min in a muffle furnace so as to stabilize the films and combustion of the undesired organic substances. All the films were prepared under the similar experimental conditions. Films prepared in this way were found to have thickness of the order of a micron.

XRD patterns of the films were obtained using X Ray Diffractometer Rigaku, Model ultima-IV, Surface morphology of the films were recorded using ZEISS electron microscope (model EV018 special edition). EDAX analysis for the films was recorded via EDS OXFORD and electrical conductivity measurement was carried out using standard two probe technique. Diffuse reflectance measurements were carried out by using Shimadzu UV–vis NIR spectrophotometer UV-3600.

3. Result and discussions

3.1. XRD studies

To check the formation of crystal structure and crystal phase, the prepared pure ZnO and La doped ZnO films ($Zn_{0.98}La_{0.02}O$) were investigated through XRD. Fig. 1(a and b) shows the XRD patterns of pure ZnO and $Zn_{0.98}La_{0.02}O$ films. The prominent peaks of the diffraction pattern are at (100), (102), (110), (103), (200), and (112) with preferred growth of grains along (100) crystallographic direction after doping with lanthanum. It is observed from the figure that diffraction peaks are shifted towards the larger diffraction angle as compared with pure ZnO. This shift may be due to the change in lattice parameter values of La doped ZnO from pure ZnO. The lattice parameters are calculated for pure ZnO are a = 3.2440, c = 5.1982 and for La doped ZnO are a = 3.2445, c = 5.1995. This increase in the lattice parameter and shift in the peaks position indicate that La^{3+} ions (1.14 Å) with larger ionic radius have been successfully incorporated in Zn^{2+} ions (0.85 Å), however it is difficult for La^{3+} ions to replace the Zn^{2+} ions as seen in case of Ce doped ZnO nanoparticles [17]. As there is no other phase is formed