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Structure, optical and electrical properties of indium tin oxide ultra thin films prepared by jet nebulizer spray pyrolysis technique

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ABSTRACT

Indium tin oxide (ITO) thin films have been prepared by jet nebulizer spray pyrolysis technique for different Sn concentrations on glass substrates. X-ray diffraction patterns reveal that all the films are polycrystalline of cubic structure with preferentially oriented along (222) plane. SEM images show that films exhibit uniform surface morphology with well-defined spherical particles. The EDX spectrum confirms the presence of In, Sn and O elements in prepared films. AFM result indicates that the surface roughness of the films is reduced as Sn doping. The optical transmittance of ITO thin films is improved from 77% to 87% in visible region and optical band gap is increased from 3.59 to 4.07 eV. Photoluminescence spectra show mainly three emissions peaks (UV, blue and green) and a shift observed in UV emission peak. The presence of functional groups and chemical bonding was analyzed by FTIR. Hall effect measurements show prepared films having n-type conductivity with low resistivity (3.9×10^{-4} Ω-cm) and high carrier concentrations (6.1×10^{20} cm^{-3}).

1. Introduction

Indium tin oxide (ITO) is a well known n-type transparent conducting oxide material. Here tin acts as a cationic dopant in the In_2O_3 lattice and as a substitute on the indium sites to bind with the interstitial oxygen. Due to its high optical transmittance, electrical conductivity and wide band gap (>3.5 eV), ITO has been widely applied in various optoelectronic devices such as photovoltaic cells [1], liquid crystal displays [2] and gas sensors [3]. The ITO thin films are commonly fabricated by employing different techniques such as magnetron sputtering [4–8], sol–gel process [9–11], thermal evaporation [12], pulsed laser deposition [13,14], chemical vapor deposition [15,16], spray pyrolysis [17–22] and nebulizer spray pyrolysis (NSP) [23]. All of these methods have advantages and disadvantages, but jet nebulizer spray pyrolysis has a noticeable advantage; it is a low-cost and non-vacuum technique for large area applications and can produce high quality film with low precursor volume. The working of NSP method is based on the Bernoulli principle; i.e., when a pressurized flow of air is directed through a constricted orifice, the velocity of the airflow is increased to create a jet stream. The impact of a jet stream with liquid produces aerosol particles (particle size ~2.5 μm) [24]. The mist form of solution is helping to improve the quality of film and to obtain a uniform growth due to gradual nucleation with minimum wastage. In the present study, the tin doped indium oxide thin films were prepared by a simple and low-cost jet nebulizer spray pyrolysis technique. The structure, surfaces, optical, photoluminescence and electrical properties of prepared films were investigated in detail.

2. Experiment

A jet nebulizer spray pyrolysis apparatus (Fig. 1) is used in this work, which consists of a jet nebulizer spraying unit, substrate holder with heater and air compressor. To prepare tin doped indium oxide thin films, the indium (III) chloride ($InCl_3$) is dissolved in 100 mL double distilled water to make 0.4 M starting solution. Tin doping was achieved by adding tin (II) chloride dihydrates ($SnCl_2{\cdot}2H_2O$) to the starting solution. A few drops of acetic acid were added to obtain a clear and homogeneous solution. The doping level in the solution was varied from 0 to 30 wt% in steps of 10 wt%. The mixture was stirred under constant speed for 1 h with a magnetic stirrer. Prior to the deposition, glass substrates (1 $in.^2$) were cleaned with acetone, isopropyl alcohol, and distilled water successively for 15 min in ultrasonicator. The substrate

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Nomenclature

D	Crystallite size (nm)
k	Scherrer's constant
d	Lattice spacing (Å)
h,k,l	Miller indices
TC	Texture coefficient
$I_{(hkl)}$	Peak intensity
N	Number of peaks
T	Transmittance (%)
$h\nu$	Photon energy (eV)
E_g	Optical band gap (eV)
n	Carrier concentrations (cm^{-3})
β	Full width at half maximum (rad)
λ	Wavelength of X-ray (Å)
θ	Bragg's angle (deg)
ε	Strain
α	Absorption coefficient
ρ	Resistivity (Ω-cm)
μ	Mobility (cm/(V s))

temperature for each deposition was kept at 500 °C in the air atmosphere. The prepared solution was sprayed with a jet nebulizer (HUDSON RCI micro mist, droplet size is ~2.7 μm) on the heated substrate with spray rate 0.5 mL/min using compressed air as a carrier gas. The nebulizer was kept at a distance of 5 cm from substrate surface.

The structural parameters of spray coated ITO films were analyzed by X-ray diffractometer (XRD) using the PANalytical system with Cu Kα_1 radiation (k = 1.54056 Å). Surface morphology and topography were carried out by the scanning electron microscope (ESEMQUANTA200, FEI-Netherlands) and atomic force microscopy (Agilent 5500) respectively. Elemental analysis was made by using energy dispersive X-ray spectroscopy (attached to SEM). The water contact angle measurement was made by using a protractor from microphotograph. The optical properties of the films were examined with a double beam spectrophotometer (Oceans optics HR2000-USA) in the UV–vis regions. The film thickness was measured by a profilometer (SJ-301 Mitutoyo). The photoluminescence (PL) spectra were recorded using a spectrofluorometer (Cary Eclipse EL08083851) with xenon arc lamp. The IR spectrum was recorded

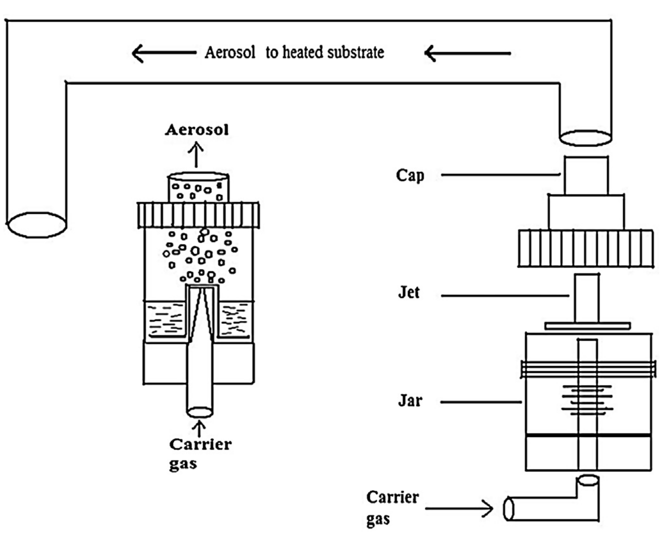

Fig. 1. Schematic diagram of jet nebulizer apparatus.

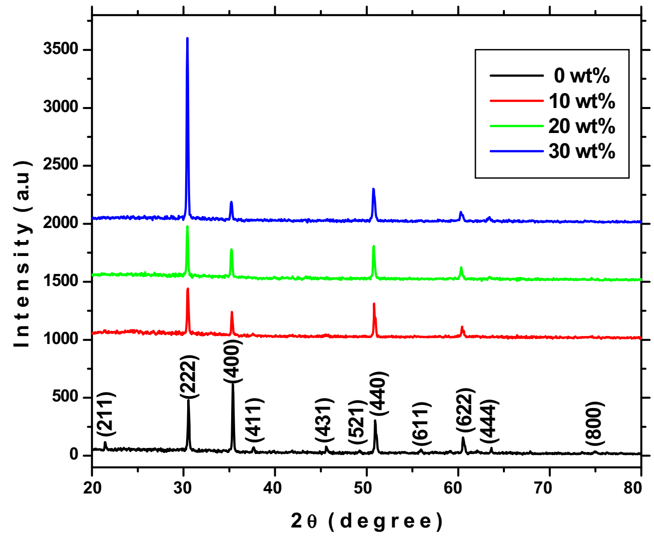

Fig. 2. X-ray diffraction patterns of ITO thin films for different Sn concentrations.

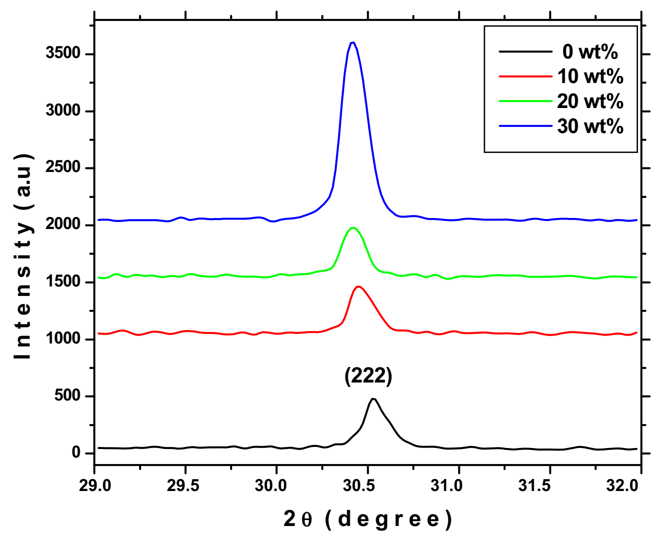

Fig. 3. Shift of peak position of ITO thin films along (222) plane for different Sn concentrations.

using FTIR spectrophotometer (Perkin Elmer – RX I) in the range of 400–4000 cm^{-1}. The electrical parameters were collected from room temperature Hall effect measurements (RH2035 PhysTech GmbH) system.

3. Results and discussion

3.1. Structural analysis

X-ray diffraction patterns are used to study the crystal structure of prepared ITO thin films. Fig. 2 shows the X-ray diffraction patterns of indium tin oxide thin films for different tin concentrations on glass substrates. It can be seen that all the films are polycrystalline in nature and crystallize in a cubic structure (JCPDS: 71-2194) with predominant (222) peak. A switching in the preferential growth from the (400) to (222) planes was observed when tin doped with indium oxide. The increasing intensity of the (222) plane is attributed to the increase in the degree of preferential crystal orientation. It is evident from the XRD spectra that no diffraction peaks of Sn or other impurity phases are detected in the prepared samples. As shown in Fig. 3 shift of the (222) peak toward smaller 2θ