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Synthesis and characterization of Aluminium and Indium co-doped Zinc Oxide thin films prepared by Pulsed Laser deposition

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ABSTRACT

Aluminium-Indium co-doped ZnO (AIZO) thin films were prepared by Pulsed Laser Deposition (PLD) technique on glass substrates. The structural, optical and surface morphology properties of the AIZO thin films were characterized by X-ray diffraction (XRD), ultraviolet-visible (UV-visible) spectroscopy and atomic force microscopy (AFM). The transmittance was found to be in the range of 81% in the visible region. XRD reveals that these films are oriented along c-axis and have hexagonal wurtzite structure. The reflection peak from the (002) plane is observed at 32.2^0 with a slight shift towards lower 2θ value as compared to undoped ZnO, suggesting enhanced out of plane lattice parameter as compared to undoped ZnO film. The surface morphology of the AIZO thin films were affected due to the presence of Aluminium and Indium. The particle size and roughness of the ZnO and aluminium-indium co doped ZnO thin film were estimated to be 33 nm, 3.8 nm and for AIZO thin films are 101 nm and 11 nm respectively.

Keywords: ZnO nanostructures thin film, indium and aluminum co-doped, optical transmission, Pulsed laser deposition

INTRODUCTION

Zinc oxide is the semiconductor compounds with a wide band gap (3.4 eV) and large exciton binding energy (60MeV) has a stable wurtzite structure with lattice spacing a = 0.325 nm, c = 0.521 nm. Transparent conductive oxide (TCO) like (SnO_2, ZnO) thin films are widely used for opto-electronic device applications, such as solar cells, flat panel displays, gas sensors, and low emissivity windows.[1-3] Nowadays, indium-tin-oxide (ITO) films are the most used TCO films for displays and solar cells in industry. However, a shortage of indium may occur in the future because of the limited nature of world indium reserves. Therefore, it is important to survey candidates like tin oxide (SnO_2) and zinc oxide (ZnO) films. Un-doped ZnO layers are highly resistive. However, the electrical properties of this material can be controlled by doping agents, usually the group-III elements (B, Al, In, Ga).[4] There are various techniques to deposit the thin films such as sol-gel process,[5] chemical vapour deposition (CVD),[6] sputtering,[7] molecular beam epitaxy (MBE),[8] spray pyrolysis,[9] pulsed laser deposition.[10,11] In this research work we have studied the effects of aluminium-indium codoped in ZnO thin films prepared by pulsed laser deposition technique.

EXPERIMENTAL DETAILS

The Aluminium-Indium co doped ZnO (AIZO) thin films were prepared using pulse laser deposition technique. The glass was used as substrates for coating. The glass substrate was rinsed with acetone and methanol each for 5 minutes. The KrF Excimer Laser was used for coating having different parameters like (λ=248nm, Laser repition rate =10Hz, pulse duration =20ns, Energy density=220 mj, deposition time =20 min. Oxygen Partial Pressure= 1 m torr.) was fixed during deposition. The prepared target was sintered at 1050 ^oC for 12 hours. The target to substrates distance was fixed to be 5 cm. The substrates temperature was fixed at 450 ^oC. The target was rotating at an angle of 94^o. The vacuum was brought at $1x10^{-6}$ torr with the help of rotary and turbo molecular pump.

The structural properties of the samples were characterized by x-ray diffraction (XRD) (Bruker D8 Advance) technology using monochromatic Cu k-α radiation to determine the crystalline structure of ZnO and AIZO (aluminium-indium co doped ZnO) thin films having wavelength (1.54056 nm). The scan rate was set in the range of 20-80^o. The morphology properties of the samples were characterized by atomic force microscopy (AFM).

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The grain size and the roughness of the samples were estimated by atomic force microscopy (AFM).The optical properties and the thickness of the following samples were measured using AMBIOUS Technology and the optical transmittance of the following samples was measured using UV-Visible spectrometer Perkin Elmer Lambda 950 in the wavelength range of the 250-850 nm.

RESULTS AND DISCUSSION

Structural properties

Figure 1 shows the x-ray diffraction of pure ZnO thin film and Al-IN codoped ZnO thin films which were deposited on the glass substrates at temperature of 450^{o}C by pulsed laser deposition technique. From figure 1 we can see that Zinc Oxide thin films (ZnO) are single phase c-axis oriented at 34.4^{o} having (002) peak and 72.1^{o} having (004) peak. The Aluminium and Indium atoms did not the affect the ZnO thin film structure,the doping concentration so small, so the diffraction peaks corresponding to Al-In doping were not observed in figure 1(b). Moreover the intensities of the peaks will be decreased due to increased in the doping concentration. The full width at half maxima (FWHM) (002) peaks also decreased due to doping. The reason is that the increase in the doping concentration deteriorates the crystallinity of the films which may be attributed to the influence of stresses ±zinc, gallium, and aluminium.

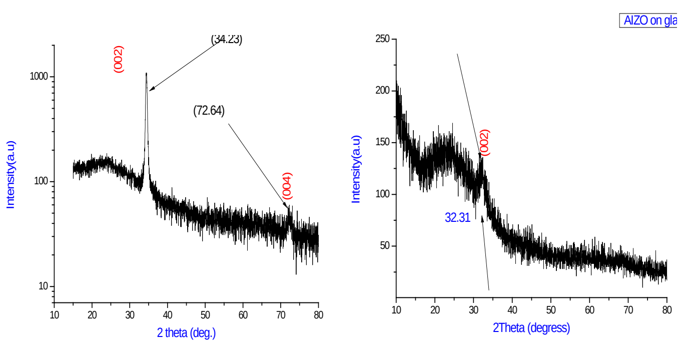

Figure 1. (a) XRD patterns of ZnO thin films (b) XRD patterns of AlZO thin films

Atomic force Microscopy (AFM)

The analysis of the ZnO and AIZO samples was done using AFM (Atomic force morphology). The scanning areas are set to be 2×2 µm. Figure 2 (a) and 2(b) shows the Atomic Force Microscopy of the ZnO in two and three dimensions and the 2(c) and 2(d) shows the Aluminium-Indium Co doped ZnO thin films in 2(c) and 2(d) by using the AFM Technology**.** The atomic force microscopy technology is used to characterize the surface morphology, surface roughness and particle size**.** The atomic force microscopy was used to determine average size of the grains nucleated in the ZnO films. The grain size and roughness of the films was to be calculated from AFM technology. The difference in the surface roughness of the doped ZnO thin films belongs to the difference in the ionic radii of the dopants that act as interstitial or substitution defects in the zno lattice. Due to the doping the grain size will increase which shows the decrease in the resistivity of the AIZO films, both the samples shows uniform surface.

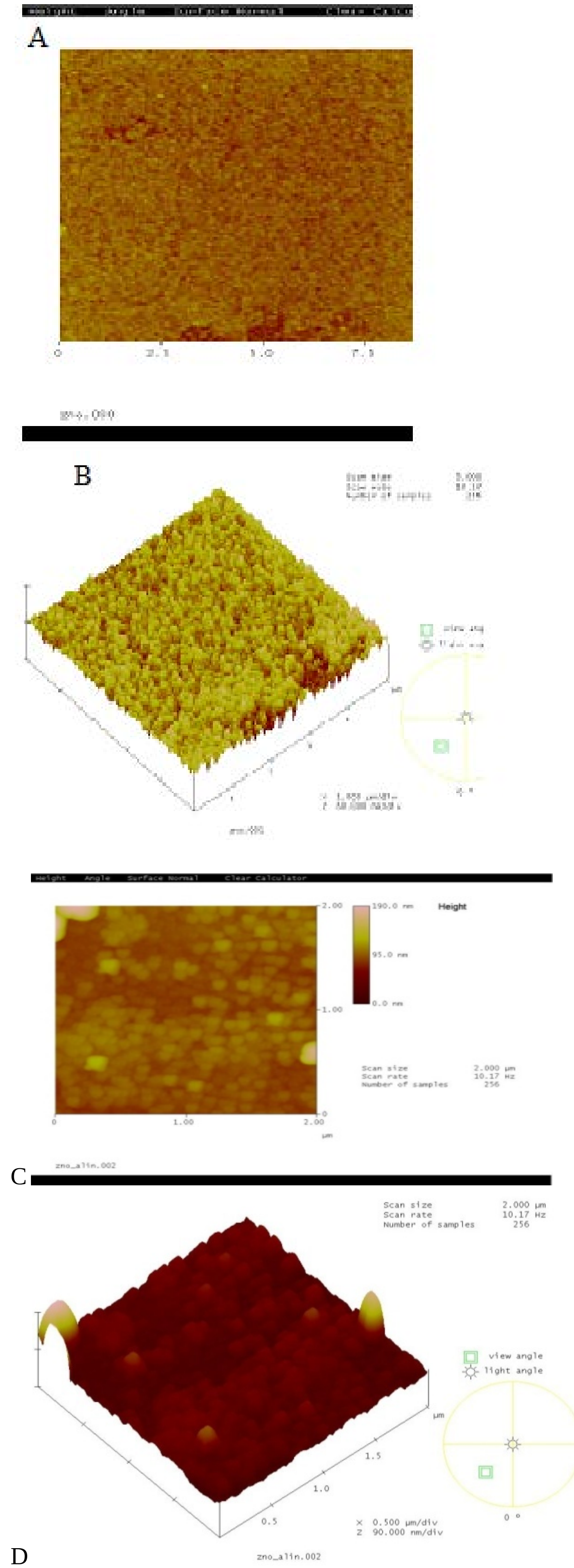

Figure 2 (a) and (b) Atomic force micrographs patterns of undoped ZnO and (c), (d) codoped ZnO thin films

The crystallite size of the ZnO and Aluminium-indium co doped Zinc Oxide thin films was calculated to be using AFM Technology 33 nm and 110nm.

Table 5: RMS roughness, Grain size of the ZnO and AIZO.

Sample	RMS roughness (nm)	Grain size (nm)
ZnO	3.9 nm	33nm
AIZO	11.3 nm	101 nm

Optical Properties

Figure 3 shows the optical transmittance of ZnO and Aluminium-Indium co doped ZnO thin films (AIZO) in the wavelength 0f 250 nm to 860 nm. The deposition temperature was kept at 450 ^{o}C for all the samples. Both the films show the transmittance in the visible region in the range of 81%. There is a sharp absorption region at around 377 nm. A UV vis.nir spectrometer is used to take the transmission of the ZnO and Aluminium-Indium co doped ZnO thin films (AIZO). It has been observed that the transmittance decreased from (92% for undoped ZnO) to 80 % due to increasing the concentration of the aluminium-indium codoped ZnO.

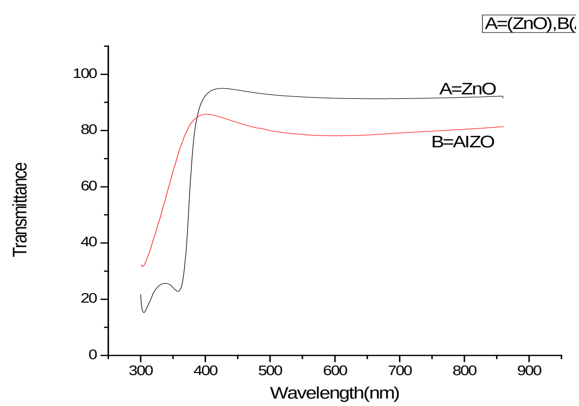

Figure 3 The transmission spectra of ZnO and AIZO.

Optical energy band gap of ZnO and AIZO

The optical energy band gap was to be calculated for undoped Zno and AIZO thin films was to be calculated to be 3.25 eV and 3.85 eV. The optical energy band gap was increased due to doping of aluminium-indium doped ZnO. A shift or a sharp absorption edge was observed and this is associated with Burstein-Moss effect.[10,12] The optical energy gap can be calculated the absorption coefficient (α) which depends on the film thickness and absorbance[13] as given in the following equations:

$$\alpha=2.303(A/d) \quad \text{--------} \ (1)$$

Where A is the absorbance and d is the film thickness. In figure 3, the transmittance will be decreased due to deformation of the crystal lattice and rough surface morphology with increasing the dopant concentration.

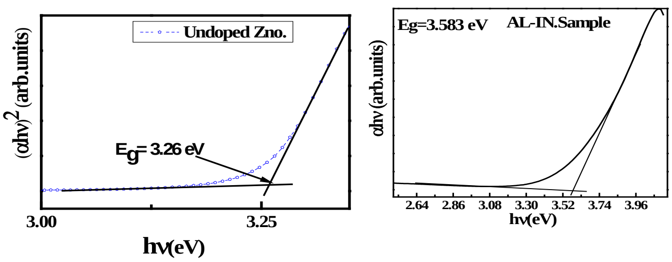

Figure 4. The optical band gap of ZnO and AIZO.

FTIR Characteristics

FTIR spectroscopy is an important technique to detect vibrational modes of synthesized samples,,semiconductor behaviours in the thin films the following samples were characterized by Fourier transform infrared spectroscopy (FTIR). Figure 5 shows the Aluminium–indium co doped ZnO thin film was taken in the transmission mode of a range of 400-4000 nm. The Characteristics was done using by using PE-Lambda Spectrometer. The Figure 4 (a) Shows the FTIR spectra of undoped ZnO thin films and aluminium indium codoped Zno thin films following peaks are observed in both the samples. The absorption peaks were observed due to the Zn-O stretching vibrations mode. Interestingly the intensity of Zn-O vibration mode will decrease due to doping of the aluminium-indium codoping with ZnO.

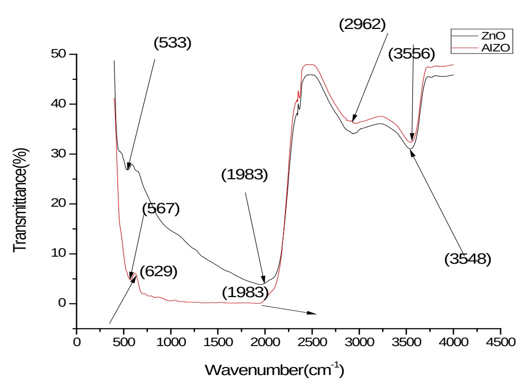

Figure 5 FTIR spectra of undoped ZnO and Aluminium-Indium co doped ZnO Thin Films.

CONCLUSION

The structural property are very well matched with Jcpds data at around 34.40 and shows a single phase behaviour and c-axis oriented axis perpendicular to the substrates surface by x-ray diffraction characterization. The optical band gap was to be calculated to be 3.22 eV. The optical properties show the transmittance of both the samples above 80 % and lies in the visible region. The AFM characteristics shows that the all grain size particles having c-axis oriented nature and the grain size and roughness for ZnO[14] and AIZO thin films varying between 33 nm to 106 nm, 3.9 nm to 4.5nm and 101nm-110 nm. The FTIR and Raman peaks also matched with the earlier data review.

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