

Formation of In₂O₃-ZnO Thin Films and Post Annealing Influence on Optical Properties

Vipin Kumar Jain, YK Vijay

ABSTRACT - In the present study, Indium-Zinc-Oxide (IZO) thin films were deposited on glass substrate with varying concentration (In₂O₃:ZnO – 90:10 and 80:20 wt.%) at room temperature using flash evaporation technique. The XRD spectra and optical micrograph show the amorphous phase of IZO thin films of vacuum thermal annealed (VTA) at 200 °C and rapid thermal annealed (RTA) for 2 minute. The optical band gap and transmittance was reduced with addition of ZnO in to In₂O₃. The effect of rapid thermal annealing (RTA) and vacuum thermal annealing (VTA) on these mixed oxide thin films also studied. The optical band gap is increased with post annealing.

Keywords: IZO thin films, XRD, Optical micrograph, Optical band gap

I. INTRODUCTION

Transparent conducting oxides (TCO) namely; indium oxide, zinc oxide, tin oxide, cadmium oxide, indium-tin-oxide, cadmium-tin-oxide etc; posses low resistivity, exhibit good adherence to many substrates, and have good transmission characteristic from visible to near infrared wavelength enabling their use in wide application [1]. During last thirty to forty years most researches have been focused on the binary compounds such as In₂O₃, ZnO, and SnO₂. There also have been rapidly growing investigations on new TCO materials involving ternary or multi component oxides such as ZnSnO₃, Cd₂SnO₄, CdSnO₃, In₂Zn₂O₅, GaInO₃ and combinations of binary or ternary oxides [2]. It is now known that doped films of oxides of tin, indium, cadmium, zinc and their various alloys exhibit high transmittance and nearly metallic conductivity is achievable. The TCO's are used in solar cells, liquid crystal display, antistatic coating on instrument panels, abrasion resistant coating, gas sensors, schottky contact to photo detectors, heat mirrors for energy efficient windows, ohmic contacts to light emitting diodes etc.[3-5] the various techniques for depositing TCO films include electron beam evaporation, ion beam sputtering, R-F magnetron sputtering, D-C magnetron sputtering, spray pyrolysis, chemical vapor deposition and thermal vacuum evaporation etc. Among multi component oxides, a ternary compound of ZnO-In₂O₃ (IZO) system proved to be very promising TCO for its good optical properties [6]. The present work deals with the study of influence of post vacuum thermal annealing (VTA) and rapid thermal annealing (RTA) on optical properties of IZO thin films were examined.

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II. EXPERIMENTAL DETAILS

IZO films were prepared by flash evaporation technique using Hind High Vacuum coating unit at a pressure of 10⁻⁶ torr. High purity In₂O₃ (99.9%)(MERC) powders and ZnO (99.99%) (M&B lab.) were mixed using mortar piston for 5 hours with varying concentration (In₂O₃:ZnO – 90:10, 80:20 wt. %). Further, a mixture of the components in powder form was fed in to a resistively heated tantalum boat by electrophonically agitating the feed chute for the instantaneous evaporation of the powdered material. The temperature of the boat was maintained at a temperature sufficiently hi

gher than the melting point of the evaporants. This high temperature ensures instantaneous vaporization of the material. The spacing of the targets to substrate was fixed at 10 cm. The typical deposition rate, deposition time and film thickness were kept constant. Thicknesses of the films were 1000 Å. The thickness was measured by using a quartz crystal thickness monitor. The substrate was kept at room temperature. The IZO thin films of different concentration were annealed by rapid thermal annealing process (RTA) for 2 minute using halogen lamp of 500 W. The similar as-deposited thin films are also annealed for 1 h in vacuum at a pressure of 10⁻⁵ torr at 473 K. The as-deposited and annealed films were characterized using XRD (Bruker AXS) with monochromatic Cu-K_α radiation (0.154nm) to identify the phase orientation. The absorption spectra of as-deposited, vacuum thermal annealed and rapid thermal annealed film were carried out in the range 290-800 nm with the help of Hitachi spectrophotometer model-330.

III. RESULTS AND DISCUSSION

X-ray diffraction (XRD) analysis

The annealed IZO thin films were characterized by X-ray diffraction (XRD) to see the nature of films. Fig. 1 (a) and Fig. 1 (b) show the XRD patterns of IZO (80:20 wt.%) thin films after vacuum thermal annealing (VTA) at 200 °C and rapid thermal annealing (RTA) for 2 minutes respectively. Fig. 1 (a) and Fig. 1(b) show the amorphous nature of the films.

Surface morphological studies

The surface morphology of the films has been studied by the optical micrographs. Fig. 2 (a) and Fig. 2 (b) show optical micrographs of vacuum thermal and rapid thermal annealed IZO (80:20 wt. %) thin films. The micrographs indicate that films are crack-free. The absence of apparent grains on the surface of both type of annealed IZO (80:20 wt. %) films can be interpreted as amorphous nature of film.

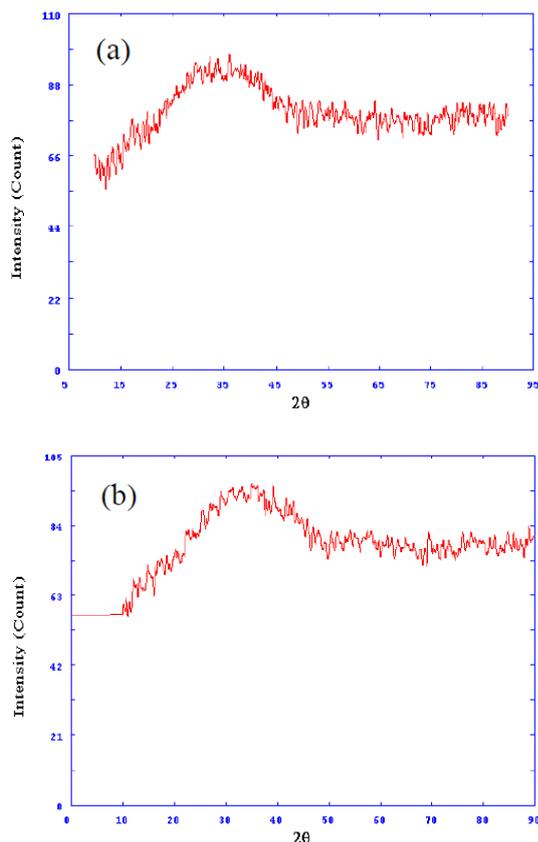


Fig 1: XRD profile of IZO (80:20 wt.%) thin films (a) after VTA at 200 °C for 1h (b) after RTA for 2 min.

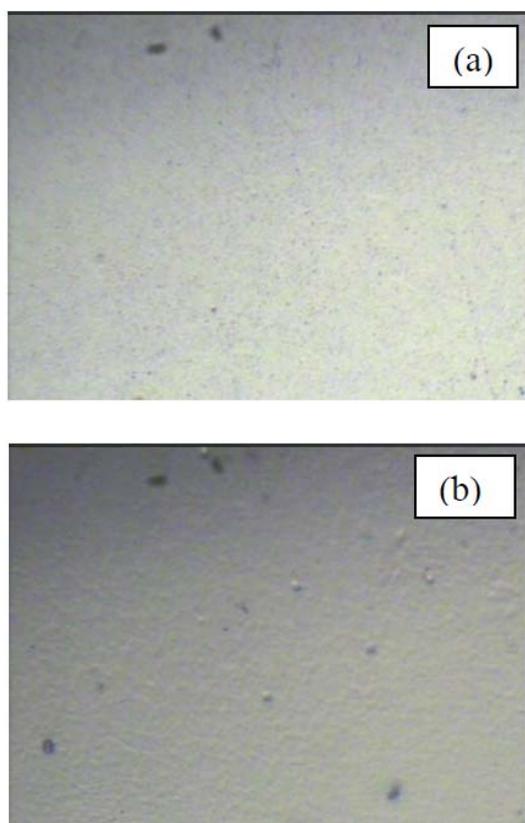


Fig 2: Optical micrograph of IZO (80:20 wt.%) thin films (a) after VTA at 200 °C for 1h (b) after RTA for 2 min.

Optical Characterizations

To determine the optical band gap, absorption spectra of as-deposited, vacuum thermal and rapid thermal annealed IZO thin films with varying concentration of ZnO was measured over the wavelength range from 300 nm to 800 nm. The optical band gap of IZO thin films can be evaluated by Tauc relation [7]

$$\alpha h\nu = A(h\nu - E_g)^n$$

where $n=1/2$ for direct allowed transition and E_g is the transition energy gap and $h\nu$ is the photon energy. Thus, for direct allowed transition $[(\alpha h\nu)^2 = A(h\nu - E_g)]$, the variation of photon energy dependence of $(\alpha h\nu)^2$ i.e. variation $(\alpha h\nu)^2$ with $h\nu$ are shown in Fig. 3 (a and b) for different composition respectively. The extrapolation of linear part of the curves towards no absorption ($\alpha h\nu = 0$) gives the effective band gap $E_{g\text{eff}}$ of IZO thin films. The values of obtained optical band gap of IZO films are given in the Table-1. It is clear from the table that the value of band gap decreases with increasing concentration of ZnO. The values of band gap are slightly higher for annealed film as compared to as-deposited films. The decrease in optical band gap with increase in ZnO concentration arises from the compositional or structural variations of the IZO films. The increase in optical band gap of IZO films after annealing may be due to the Burstein-Moss shift (BM shift). The shift occurs owing to blocking of the lowest conduction band states by partial filling of electrons above the mott critical density. The shift is given by the relation,

$$E_g = E_{g0} + \Delta E_{g\text{BM}}$$

Where E_{g0} is the intrinsic band gap of IZO (when the carrier density is zero) and $\Delta E_{g\text{BM}}$ is the BM shift [8].

It is also observed from the figure 4 (a and b) that the transmittance of the IZO films is improved after annealing and decreased with increasing concentration of ZnO. It may be due to presence of mixed oxide phase and oxygen vacancies.

Table 1: Optical band gap of IZO thin films

| Specimen | as-deposited | VTA (200°C) | RTA (2 min.) |
|-------------------|--------------|-------------|--------------|
| IZO (90:10 wt. %) | 3.99 eV | 4.01 eV | 4.04 eV |
| IZO (80:20 wt. %) | 3.92 eV | 3.97 eV | 4.00 eV |

IV. CONCLUSION

It is concluded from the above study that as-deposited, post vacuum annealed at 200 °C and rapid thermal annealed IZO thin films were amorphous in nature. The addition of ZnO in to In_2O_3 reduces the transmittance and optical band gap of the IZO thin films. The transmittance and optical band gap were improved after annealing.

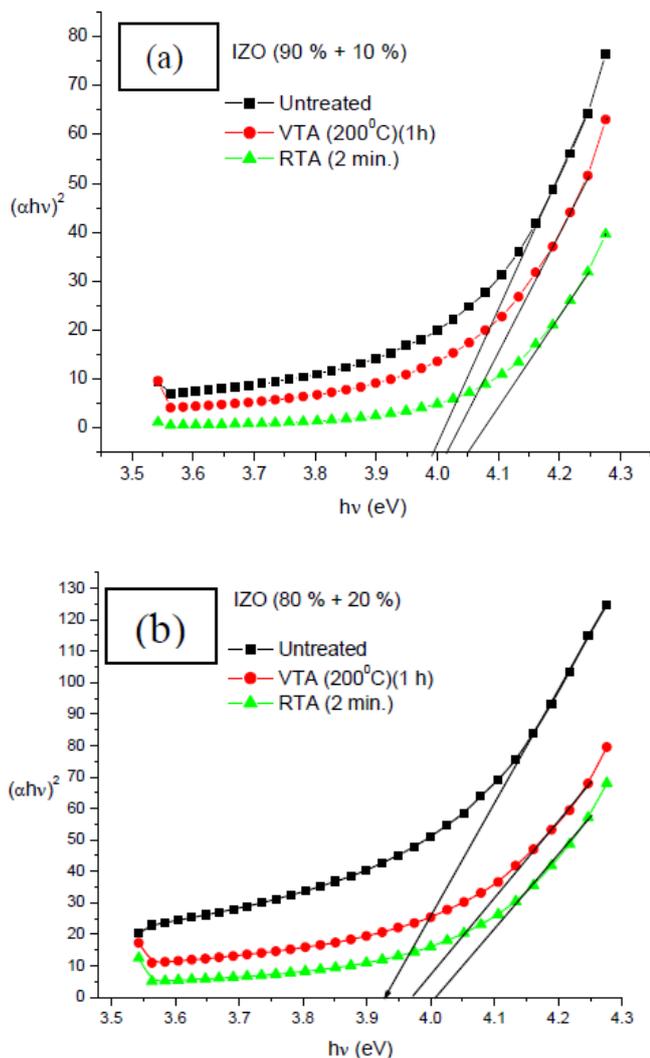


Fig 3: Optical band gap of (a) IZO (90:10 wt.%) and (b) IZO (80:20 wt.%) thin films for untreated, post VTA at 200 0C for 1h and RTA for 2 min

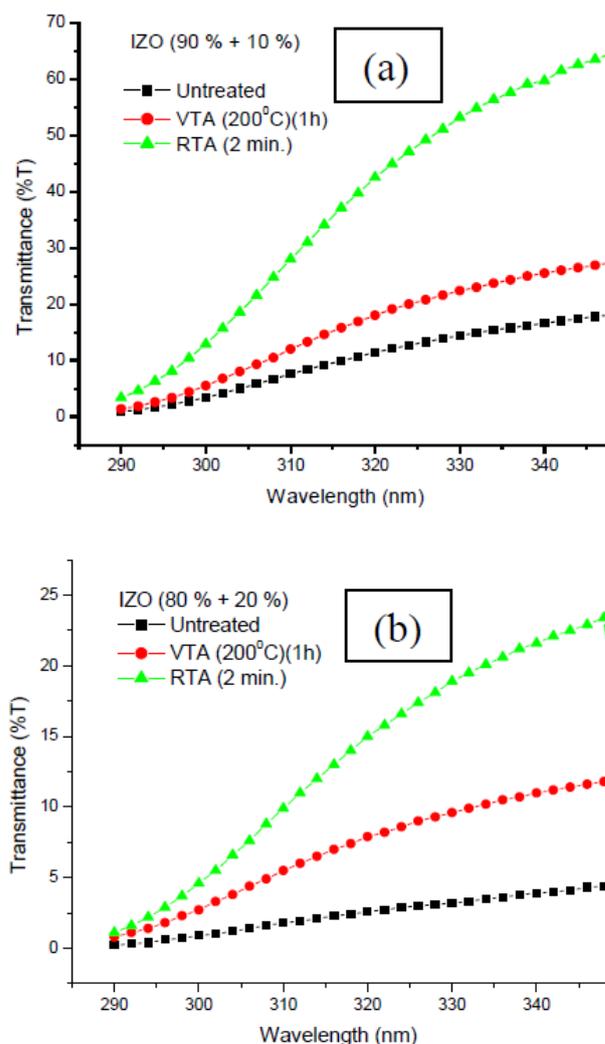


Fig 4: Optical transmission spectra of (a) IZO (90:10 wt.%) and (b) IZO (80:20 wt.%) thin films for untreated, post VTA at 200 0C for 1h and RTA for 2 min. **Dsfsdf**

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