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REVIEW ARTICLE

STRUCTURAL AND OPTICAL PROPERTIES OF ZnO THIN FILMS PREPARED BY SPRAY PYROLYSIS FOR DIFFERENT SUBSTRATE TEMPERATURES

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ABSTRACT

ZnO thin films can be grown by various techniques implemented by various temperatures. We present the Morphological and Optical properties of ZnO thin films prepared by using spray pyrolysis on glass substrates at various growth temperatures from 150°C, 200°C, 250°C. The surface morphology of the films analysed by scanning electron microscopy SEM. The optical energy gap for all thin films was calculated using Tauc's equation. The band gap of ZnO films increases with increasing substrate temperatures attributed to the increase of the grain size of the sample. The calculated band gap energy of ZnO thin films deposited at 150°C, 200°C, 250°C varies from 3.2eV to 3.32eV, which correspond to a slightly change in the band gap energy with the substrate temperature. From X-ray diffractometry measurements, zinc oxide thin films showed hexagonal wurtzite structure.

INTRODUCTION

Transparent conducting zinc oxide has attracted considerable attention on account of its good electrical and optical properties in combination with its abundance in nature and non-toxicity. Zinc oxide has become an promising material for many applications in solar cells (Desai *et al.,* 2012), gas sensors (Mitra *et al.,* 1998), optics, microelectronic devices (Jackson *et al.,* 2005) and sensing because of its direct wide-band gap of 3.37eV, n-type nature and large binding energy of 60 meV at room temperature (Fenollosa *et al.,* 2005; Ding, *et al.,* 2009). Is is also attractive in the fields of semiconductor because of its optical properties, high stability, cheap, and good electrical properties compared to other materials. The electrical resistivity of ZnO is modified by the reactive deposition. Other advantages of the use of ZnO layers in photovoltaic cells concern its high chemical and physical stability, thermal stability in hydrogen plasma atmosphere, high electrical conductivity and finally, its low cost price. ZnO has been fabricated by using various methods, such as pulsed laser deposition (Jin *et al.,* 2000), sol-gel (Khan *et al.,* 2011), rf-sputtering (Minami *et al.*), chemical vapour deposition (Khusaimi *et al.,* 2009), electrochemical depositon (Yoshida *et al.,* 2004), molecular beam epitaxy (Heo *et al.,* 2008), and spray pyrolysis (Ayouchi *et al.,* 2003). Among all the methods mentioned, spray pyrolysis is one of the effective and attractive techniques for preparing ZnO thin film on the in expensive substrate. This is because of simplicity, vaccum free technique, low cost and large film area (Ayouchi *et al.,* 2003). Some of the factors that may affect the physical properties and the quality of the film in spray pyrolysis process include the spray temperature, precursor concentration, the rate of spray, spraying geometry and distance between nozzle and the substrate (Rajalakshmi *et al.,* 2011). By spray pyrolysis, a large number of materials were deposited namely SnO2, Cds, CdTe. The used precursor solutions are generally zinc acetate (Van Heerrden, 1997; Nunes *et al.,* 2001), zinc nitrate (Studenkin *et al.,* 2008) and zinc chloride (Eberspacher *et al.,* 1986) while the mostly used one is the zinc acetate. In the present paper we study the influence of the solution and properties on the characterization of ZnO thin films deposited by spray pyrolysis techniques.

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Experimental Details: The ZnO thin films were formed by the spray pyrolysis techniques. In this deposition technique, a solution of zinc acetate dehydrated (Zn (CH3COO)22H2O) with 0.1 M is used as the precursor solution, prepared by dissolving in a mixture of deionised water. A few drops of acetic acid were added to the aqueous solution to prevent the formation of hydroxides. The nozzle was a distance of 25cm from the substrate during the deposition. The solution flow rate was held constant at 3ml/min. Air was used as a carrier gas at the pressure of 2 bar. The fine droplets of the prepared solution were sprayed onto the hot substrate where thermal decomposition of the reactants in the precursor solution occurred, leading to the formation of ZnO layers on the glass substrates. Thus, the chemical in the solution are vaporize react on the substrate surface after reaching it and ZnO thin films are deposited the glass substrate. The ZnO thin films were deposited at substrate temperatures of 150^{o}C, 200^{o}C, 250^{o}C.

RESULTS AND DISCUSSION

A. Structural Characterization: The Crystalline state of the films for the various substrate temperature has been studied using XRD.

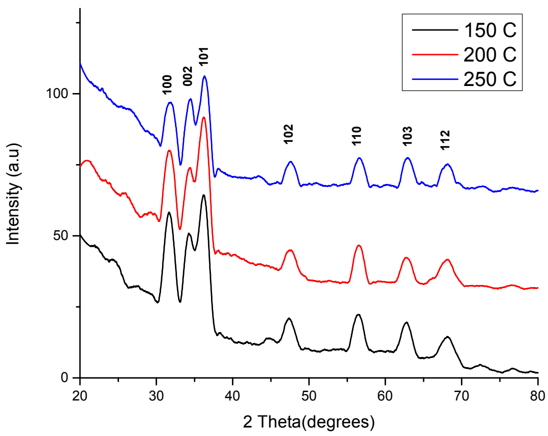

Fig. 1. XRD pattern of ZnO thin films for various temperatures.

Fig.1 shows the diffraction patterns of the films grown under 2 bar air pressure. ZnO is detected for 150^{o}C, 200^{o}C, 250^{o}C temperatures. The four well defined peaks, (100), (002),(101), (110) diffraction planes of ZnO, are noted in the diagrams indicating the polycrystalline wurtize structure of ZnO. And the intensity of the (002) peak detected around 34.36^o. The films have random growth orientation. This is because of the amophorous nature of the substrates for various temperatures. The mismatch between the polycrystalline film and the amphorous substrate may occur the strains in the thin films. For the hexagonal crystals, the stress in the plane of the film is calculated by using the biaxial strain model. The total stress in the films consists of two components. One is the intrinstic stress occur due to the impurities and lattice distortion in the crystals and other is the extrinstic stress occur due to thermal expansion coefficient mismatch between the film and the substrate. According to the srtain equation,if the stress is negative, the film is compressive and if the stress is positive, the film become tensile. The average crystallite size of the films deposited at various substrate temperatures have been calculated using the Scherrer's formula,

$$D = \frac{0.9\lambda}{\beta cos\theta}$$

where,
λ, β , θ are x-ray wavelengths, the Bragg's diffraction angle and the full width at half maxima (FWHM) of the peak corresponding to θ value respectively.

B. UV-vis-NIR data analysis: A high optical transmittance in the visible range is required for most of the applications of ZnO thin films. Fig.2 shows the energy band gap (E_g) of the grown temperature at various temperatures (150^{o}C, 200^{o}C, 250^{o}C). The energy band gap (E_g) of the as grown films can be determined by applying the Tauc model by extrapolating the linear part of the straight lines onto the energy axis to reach (αhϑ) =0.

The band gaps for different temperatures are 3.2 eV, 3.22 e V, 3.32 e V respectively. The compressive stress in the films causes the increase of band gap. The compressed lattice is expected to provide a wider band gap because of the increase repulsion between the oxygen 2p and the zinc 4s bands.

C. Microstructure: The substrate dependence of the crystal quality and the crystal morphology in the ZnO thin flims is further analysed by SEM images of ZnO flms for various temperature 150^{o}C, 200^{o}C, 250^{o}C and is shown in the Fig. 3(a), 3(b) and 3(c) respectively. The morphology of 150^{o}C sample is formed of, many size agglromated particle distributed on the suface of the film. For, 200^{o}C the particles are small crystals and are evenly distributed over the films and for 250^{o}C, the particle is grain size and is found to be tiny and homogeneous in structure. It can be concluded that, as the temperature increases the particle of the grain size decreases.

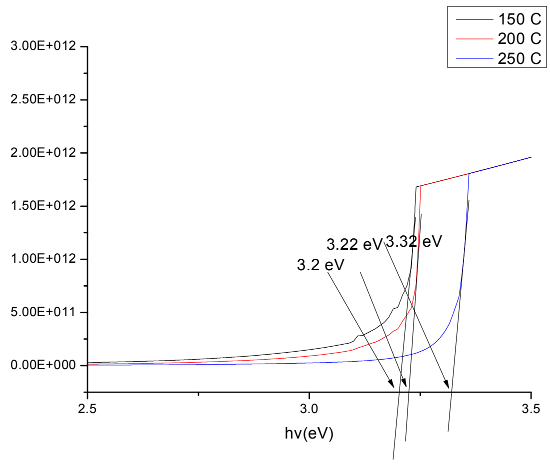

Fig. 2. Variation of $(\alpha h\vartheta)^2$ vs hv of the ZnO thin films.

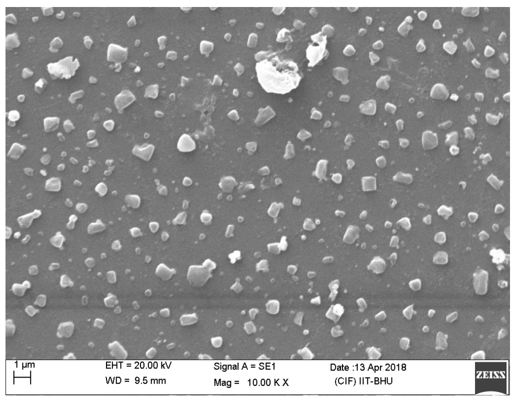

Fig. 3(a) 150^{o}C

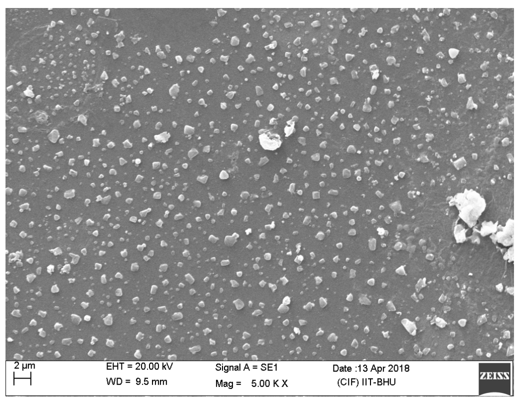

Fig. 3(b) 200^{o}C

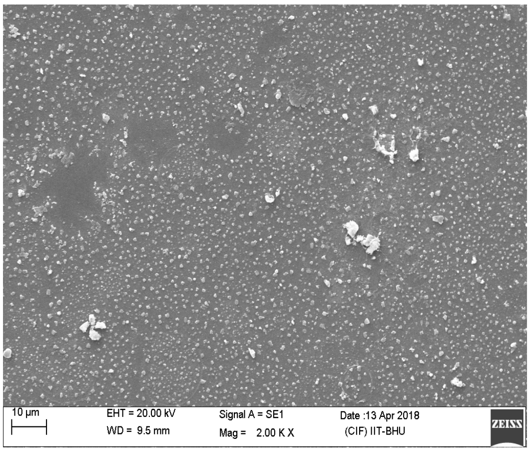

Fig. 3(c) 250^{o}C

Conclusion

In this study, the influence of the substrate temperature on the structural, optical properties of ZnO thin films grown on the glass substrates by spray pyrolysis method was investigated. The XRD studies show that the films deposited at low temperature have large stress. SEM analysis revealed that films at the different temperature changes its grain size agglomeration particle and distributed all over the substrate of the thin films. The optical analysis exhibited the change in the energy gap from 3.2 e V, 3.22 e V, and 3.32 e V for 150^{o}C, 200^{o}C, 250^{o}C.

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