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STRUCTURAL AND MORPHOLOGICAL CHARACTERISTICS OF ZINC OXIDE THIN FILMS PREPARED BY SPRAY PYROLYSIS TECHNIQUE

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ABSTRACT

ZnO thin films have been deposited on the glass substrates by spray pyrolysis method. In this work we have studied the structural and morphological properties of ZnO thin films. Zinc acetate dihydrate was used as the precursor material. The structural and morphological properties of the films have been investigated by X-ray diffraction and atomic force microscopy characterization methods. Scherrer's formula was used to calculate particle size. The thin films have (002) as the preferred orientation. The root mean square roughness of the films was calculated.

KEYWORDS: Zinc oxide film, spray pyrolysis, XRD, AFM

I. INTRODUCTION

Thin zinc oxide (ZnO) films are of great importance due to their unique optical and physical properties. ZnO with its extraordinary properties have a very promising semiconducting material used in many branches. ZnO belongs to the group AII-BVI semiconductors and it has composed of hexagonal wurtzite crystal structure. ZnO thin film presents investigating structural, morphological properties which meet extent applications in the fields of electronics, optoelectronics and sensors. ZnO thin film is applied to the transparent conductive film and the solar cell window because of the high optical transmittance in the visible region[1]. It reveals the zinc oxide has a wide band gap of 3.3 eV and largest exciton binding energy (60 mV) among the members of this group [2-5]. Zinc oxide is an n-type semiconductor with very important properties, which make it commonly used in electronic and optoelectronic applications [6],optics [7, 8], where the zinc oxide is used as a component for UV and blue range, and also as an optical waveguide [9], photocatalyst [10], or solar cells window [11-13]. Furthermore the manufacture of zinc oxide in a form of piezoelectric transducers, varistors, phosphorescent substances, transparent conductive films, transistors TFT (thin film transistors) or gas sensors has been also reported [14-16].
Several methods are applied to prepare ZnO films, both physical and chemical deposition technologies including sputtering [17], pulsed laser deposition [18], chemical vapor deposition (CVD) [19], molecular beam epitaxy [20], and sol–gel process [21] etc. Spray pyrolysis is a useful alternative to the traditional methods for obtaining thin films of ZnO. It is of particular interest because of its simplicity, low cost and minimal waste production. In spite of few studies regarding to the spray pyrolysis method, the spray pyrolysis method has some merits, such as the easy control of chemical components. These techniques have distinct advantages such as easy control of chemical composition, simplicity minimal waste production, low cost and it is also non-toxic and environment friendly.

In the present work, thin films of zinc oxide work prepared using spray pyrolysis method and to study the structural and morphological properties of ZnO thin films as functions of deposition temperature and pressure, liquid precursor flow, carrier gas flow and the inert and reactive gas ratio along with micro structural quantities.

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II. MATERIALS AND METHODS

ZnO thin films were prepared on glass substrates (76×26×1.5mm) by spray pyrolysis method. The glass substrates were cleaned with soap solution followed by acetone. Then it was degreased in acetone and ethanol 1:1 for final cleaning. The deposition solutions were formed by first dissolving weighed amounts of 0.2M solution of zinc acetate dehydrate $Zn(CH_3COO)_2 \cdot 2H_2O$ diluted in methanol and deionizer water (3:1) was used for all the films. A few drops of acetic acid were added to improve the clarity of solution. Nitrogen was used as the carrier gas, pressure at 0.2 kg cm^{-2}. The ultrasonic nozzle to substrate temperature was measured using an Iron-Constant and thermocouple. In this case temperature was 325°C. The microscopic slide of borosil glass(9100P01) was used as a substrate. The starting solution was mixed thoroughly and final solution was sprayed. The nozzle–substrate separation used was of 30 cm. During the spraying process, the substrates were heated by electrically heating the copper plate.

$$Zn(CH_3COO)_{2(\text{solid near substrate})} \rightarrow 4Zn(CH_3COO)_{2\,(\text{gas near substrate})}$$
$$4Zn(CH3COO)_2 + H_2O \rightarrow Zn_4(CH3COO)_6 + 2CH_3COOH_{(\text{gas near substrate})}$$
$$Zn_4(CH_3COO)_6 \uparrow +3H_2O \rightarrow 4ZnO_{(\text{film})} + 6CH_3COOH_{(\text{gas})}$$

The color of the deposited ZnO thin films was milky white. In the present work the deposition time was one hours. The structural properties of the prepared films were studied by X-ray diffraction technique (Bruker D8 Advanced Diffractometer (λ= 1.54059 Å)).The surface morphology was investigated using atomic force microscope (AFM, PARK XE-7).

III. RESULTS AND DISCUSSION

Structural Properties

The crystallographic structure of the ZnO films was examined by powder X-ray diffraction technique. An x-ray diffraction spectrum of the ZnO thin film is shown in Fig. 1. An x-ray diffraction spectrum of all the films was taken at room temperature. The peaks with the Miller indices given belong to the ZnO [22]. The presence of sharp structural peaks in these X-ray diffraction patterns confirmed the polycrystalline nature of the films. As shown in Fig. 1, the thin film have (0 0 2) for the preferred orientation. This (0 0 2) preferred orientation is due to the minimal surface energy for which the hexagonal structure, *c*-plane to the ZnO crystallites, it also corresponds to the densest packed plane [23]. This result is in good agreement with literature data [24–27].
The particle size for the samples was calculated from the X-ray diffraction data by using Scherer's formula.

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

Where, λ = wavelength of the X- rays (1.5406Å), β= FWHM of the peak with highest intensity and θ = diffraction angle.

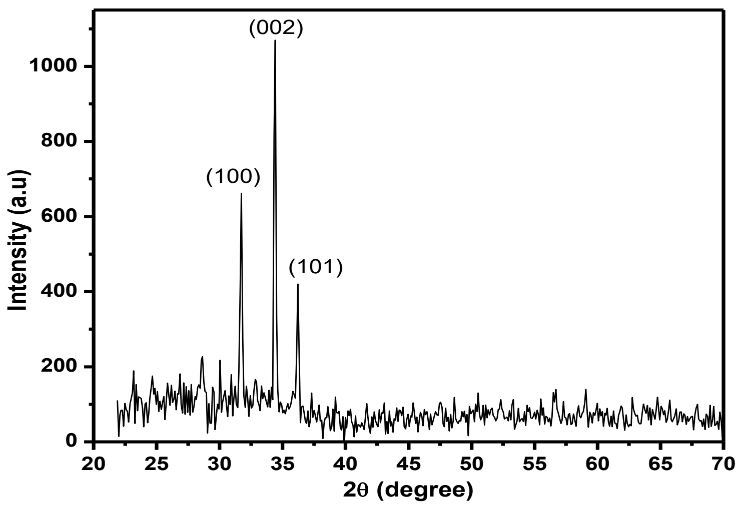

Figure 1. XRD pattern of ZnO thin film deposited at 325°C

From the standard equations the particle size, dislocation density, micro strain and lattice constant of ZnO thin films deposited at 325°C were calculated and presented in Table 1.

Morphological Properties

The AFM analysis allowed us to study the morphology of the film produced, to infer the size of the grains formed, and to analyze possible defects existing between grains (Fig.2). The data achieved reveal films with different surface morphologies and grain sizes. The surface images obtained by AFM, showing spray pyrolysis films with a high surface roughness is 73 nm and high grain sizes 40 nm.

The AFM image of the film shows porous structure consisting of grains with different sizes separated by empty spaces. Atomic force microscopy was used to examine surface roughness of the films over a cross-sectional area of 5 µm x5 µm as shown in Fig.2.

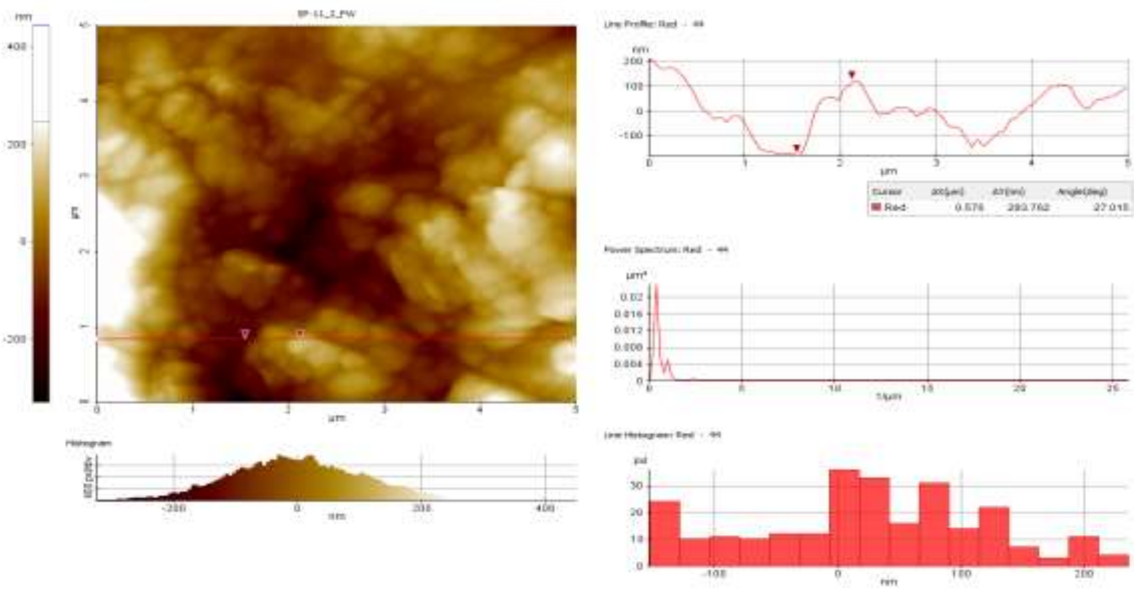

Figure 2.AFM image of ZnO thin film deposited at 325°C

Table 1. Micro structural quantities of ZnO thin films

2θ	Particle Size (nm) D	Dislocation density (δ)line^2/m^2 x10^{15}	Micro strain (ε) x10^{-3}	Lattice constant (A^o)	
				a	c
31.732	40.1933	0.619	0.862	3.6684	5.6330
34.4142	40.5130	0.609	0.855	3.2526	5.2057
36.2061	36.9189	0.734	0.939	3.0135	4.9561

IV. CONCLUSION

Zinc oxide thin films have been successfully deposited on glass substrates by the spray pyrolysis deposition technique. From the X-ray diffraction analysis, the deposited film show hexagonal structure along with c-axis oriented (002) plane and average particle size is 40 nm. Morphology of the ZnO thin film shows high roughness and larger grains. The root mean square roughness (Rq) of the ZnO thin film is nearly equal to 73 nm.

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