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Impact of Thickness on Nano-Structured ZnO Thin Films Fabricated by Spin Coating Method

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Abstract: *In this work, Nano-Structured ZnO thin films were prepared by Spin coating technique on glass substrates at various layers. The structural and optical properties were characterized by atomic force microscopy measurement (AFM), field emission scanning electron microscopy (FESEM) and UV-Vis-NIR respectively. The surface morphology reveals that the nano-structured ZnO thin films become densely packed as the thickness increases. The average grain size of nano-structured ZnO thin films varied from 18nm to 25nm with an increase in thickness as well as increase of the surface roughness. All the films are transparent in the visible region (400-800 nm) with average transmittance above 85 %.*

Keywords: *Zinc Oxide, Spin Coating, Structural Properties, Optical Properties*

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I. INTRODUCTION

ZnO is one of the most important wide-band gap semiconductors that are beneficial for optoelectronic devices application. It has a tunable physicochemical property, high surface activities, economical and non-toxicity [1]. These advantages make it useful for photocatalysis, purification air and water [2], field-effect transistors [3], dye sensitized solar cells [4], gas sensors [5], etc.

Zinc Oxide, a well-known II-IV compound semiconductor has a direct band gap of 3.37eV and 60meV of free-exciton excitation energy at room temperature and it useful for dye-sensitized solar cell [6]. Higher electron mobility in ZnO thin films will reduce the electron recombination and the electron easily injected to the conduction band thus will enhance the performance of solar cell [7]. Many researchers reported that the performance of solar cells is directly related to the good crystalline structure of ZnO thin film [8-9]. The ZnO thin film that has good crystalline structure has been used to grow a variety of structures such as nanowires, nanorods, etc. In dye sensitized solar cells it has been applied as a seed layer for nanostructures growth as well as for buffer or blocking layer. There are several methods that have been used to synthesize ZnO thin films: chemical vapor deposition (CVD) [10], molecular beam expitaxy (MBE) [11], radiofrequency (rf) magnetron sputtering [12], sol-gel [13]. Among these, sol-gel has advantages such as good uniformity of thin film, high purity, and low temperature synthesis and toward cost-effective production [14-15]. In device applications, the enhancements of thin films could affect the performance of devices.

In this paper, we report the effect of the various layers on the structural and optical properties of Nano-Structured ZnO thin films deposited on glass substrates by spin coating method. The properties of the nano-structured ZnO films deposited at different deposition times are systemically investigated by field emission scanning electron microscopy (FESEM), atomic force miroscopy (AFM) and UV-Vis-NIR spectrophotometer.

II. EXPERIMENTAL

Microscope glass was used as substrates were cleaned in the acetone, methanol and deionised water using HWANSHIN Powersonic 405 ultrasonic cleaner in order to remove all the contamination. Then, the nitrogen gas was used to dry the cleaned glass substrates before the deposition process by Sol gel spin coating techniques.

Nano-strcutured ZnO thin films were prepared using 0.4M zinc acetate dihydrate as a precursor, 2-methoxyethanol as a solvent and monoethanolamine (MEA) as a stabilizer with molar ratio 1:1. The solution was stirred and heated for 3 hours before aged for 24 hours at room ambient. Then, the solution was spin-coated on glass substrates at a speed of 3000 rpm for 1 minute. The as-deposited films were heat-treated at 150 °C for

10 minutes in order to evaporate the solvent. The coating procedures were repeated for 1, 3, 5, 7 and 9 times to increase the film thicknesses and represented as 1 layer, 3 layers, 5 layers, 7 layers and 9 layers. After the coating process, the thin films were annealed for 1 hour at 500 °C in ambient air.

The film thicknesses were measured using the Veeco/D 150+ surface profiler. Meanwhile, the structural properties were characterized by field emission scanning electron microscopy (FESEM, JOEL) and atomic force microscopy (AFM). The optical properties were obtained by using UV-Vis-NIR spectrophotometer (Jasco V- 670).

III. RESULT AND DISCUSSION

A. Surface Morphology

Figure 1 illustrates the pictures and average grain size of the nano-structured ZnO thin films produced on glass substrates for deposition durations of 1, 3, 5, 7, and 9 minutes. FESEM images indicate that the morphologies of the nano-structured ZnO thin films become denser with higher deposition across various layers. Table I presents the thicknesses of nano-structured ZnO thin films across several layers. The FESEM pictures revealed the homogeneity and crystallinity of all ZnO thin films, consistent with previous reports [16-17]. Nevertheless, when the thickness rose, the thin film exhibited a somewhat agglomerated particle. In Fig. 1(c), the proximity of particle configurations is significant for establishing routes in the granular layer to facilitate electron flow. Conversely, other ZnO thin films exhibit closely positioned particles, while some pores and agglomerated particles are present.

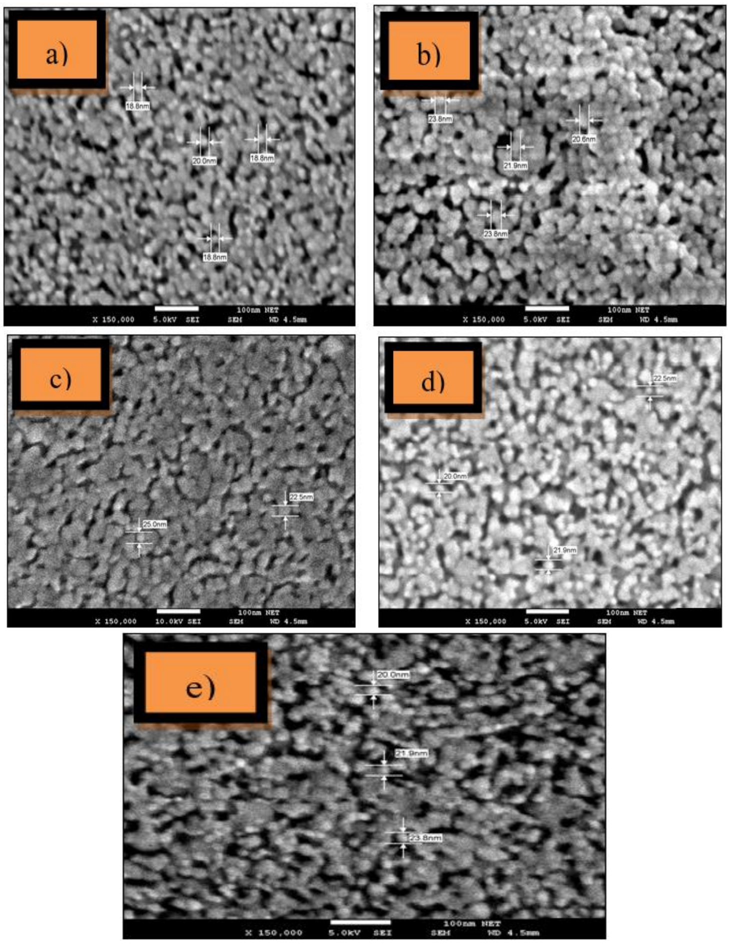

Fig. 1 FESEM images depicting the measured grain size of ZnO thin films at (a) 1 Layer (b), 3 Layer (c), 5 Layer, and 9 Layer with a magnification of 150,000 and 5.0 kV

TABLE I
THICKNESS OF NANO-STRUCTURED ZnO AT DIFFERENT LAYERS

Number of Layers	Thickness (nm)
1	40.38
3	115.2
5	169.23
7	224.30
9	311.33

Figure 2 displays AFM pictures of the structure of nanostructured ZnO thin films at various levels, fabricated using the sol-gel spin coating process. As the thickness rose, the average roughness of the ZnO film similarly escalated. Table II indicates that the average roughness Ra of the formed ZnO thin film at varying deposition durations (1 layer, 3 layers, 5 layers, 7 layers, and 9 layers) were 0.391 nm, 0.395 nm, 0.457 nm, 0.506 nm, and 0.641 nm, respectively. This behavior has also been documented by others [18]. The elevated surface roughness of thin films, which diminishes optical efficiency, may result from the augmented scattering of incident light. The increase in average roughness contributed to the decrease in transmittance of thin films.

TABLE II
AVERAGE ROUGHESS OF NANO-STRUCTURED ZnO AT DIFFERENT LAYERS

Number of Layers	Average Roughness Ra (nm)
1	0.391
3	0.395
5	0.457
7	0.506
9	0.641

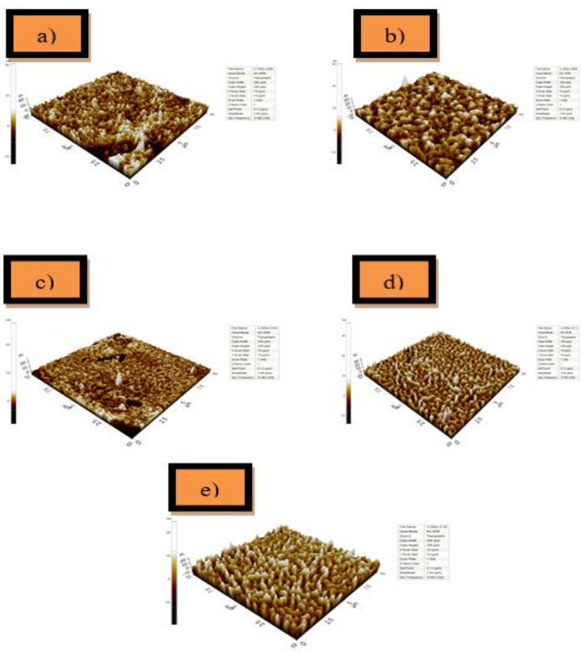

Fig.2 AFM images of ZnO thin films at (a) 1 Layer, (b) 3 Layer
(c) 5 Layer (d) 7 Layer and (e) 9 Layer

B. Optical Properties

Figure 3 illustrates the transmittance spectra of nano-structured ZnO thin films across various layer configurations. All the thin films had high transparency ranging from 80% to 96% within the visible spectrum of 400 nm to 800 nm. The thin films permit a significant fraction of the light spectrum to enter within the visible wavelength range, which may be advantageous for solar cell applications [19].

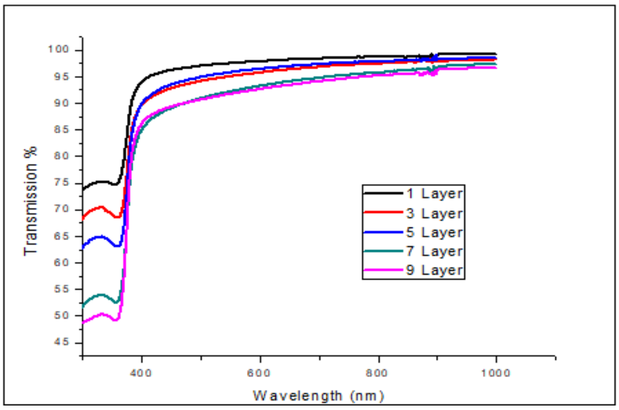

Fig.3 Transmission spectra of ZnO thin films at different layers

Figure 3 illustrates the wavelength dependency of the optical transmittance spectra for nano-structured ZnO thin films produced in varying thicknesses, as measured by a UV-Vis-NIR spectrophotometer. The optical transmission spectrum of nano-structured ZnO thin films revealed that the best transmittance occurred in the sample deposited with a single layer, whereas the lowest transmittance was noted in films deposited with nine layers. The optical transmittance of the nano-structured ZnO thin film for 3 layers, 5 layers, and 7 layers was 90-93%, 90-94%, and 87-92%, respectively, across the visible wavelength range of 400-800 nm. In the meantime, the transmittance observed in the three-layer and five-layer configurations exhibited modest variations, potentially attributable to the homogeneity and reduced grain boundaries of the five-layer nano-structured thin film, which resulted in enhanced light transmittance in the visible spectrum [20-21]. It may be argued that the transmittance of thin films diminishes as their thickness increases, yet all thin films exhibit high transmittance due to their uniformity. Table II indicates that the roughness of thin films is minimal, suggesting uniformity in their structure.

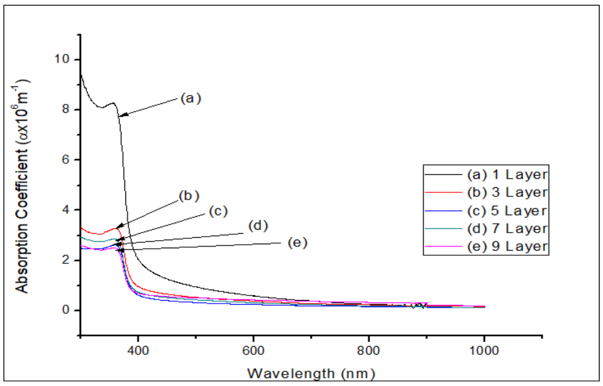

Fig.4 Absorption coefficient of ZnO thin films at different layers

The transmittance measurements will determine its absorption characteristics. Figure 4 illustrates the absorbance coefficient of nano-structured ZnO thin films deposited in varying thicknesses. The absorption coefficients were computed from transmittance data utilizing Lambert's law, as demonstrated in equation (1) [22]:

$$\alpha = \frac{1}{t}\ln\left(\frac{1}{T}\right) \tag{1}$$

where t denotes the thickness of the thin film, α represents the absorption coefficient, and T indicates the transmittance value at the corresponding wavelength. The results indicate that all films exhibit minimal absorption in the visible and near-infrared (NIR) ranges, however in the ultraviolet (UV) range, all films display substantial absorption. A significant absorption coefficient in the UV range near the band edge, around 370 nm, may result from electrons in thin films possessing sufficient energy to transition from the valence band to the conduction band when exposed to photon energy from light [23]. The optical band gap values of nano-structured ZnO were derived from transmission measurements by graphing $(\alpha h\nu)^2$ against photon energy, where α represents the absorption coefficient and hv denotes the photon energy. Figure 5 illustrates that the optical energy band gaps diminished from 3.228 eV to 3.11 eV for nano-structured ZnO thin films placed in layers of 1, 3, 5, 7, and 9, respectively. M. Ali Yildrin et al. reported that the band gap energies diminished with the increase in film thickness. This phenomenon may result from the growth process, wherein certain impurities (such as oxygen vacancies and/or Zn interstitials) establish energy levels at the conduction band edge that interact with the conduction band as thickness increases. Additionally, it may be attributed to structural defects in the thin films arising from their preparation at room temperature [24].

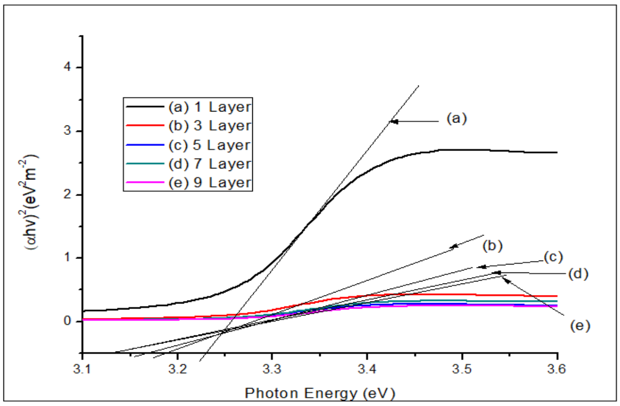

Figure 5: Estimation of optical band gap energy via Tauc's figure

IV. CONCLUSION

The impact of thickness on the structural and optical characteristics of nano-structured ZnO thin films was examined using FESEM, AFM, and UV-Vis-NIR techniques. The surface morphology indicates that the nano-structured ZnO thin films grow increasingly dense with the addition of layers. The average grain size of nano-structured ZnO thin films ranged from 18 nm to 25 nm, correlating with an increase in deposition time and surface roughness. All the films exhibit transparency in the visible spectrum (400-800 nm) with an average transmittance over 85%. The optical band gap appears to diminish with increasing thickness, measuring 3.11 eV at a thickness of 311.33 nm.

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