Effect of thickness on the physical characterization of sprayed ZnO thin films

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ZnO thin films having different thicknesses (300, 400 and 500) nm were deposited by spray pyrolysis method (SPM). XRD analysis indicate that the deposited films have hexagonal wurtzite structure and display a strong peak at (002) plane. The effects of thicknesses on crystallite size, stress and strain are investigated. The thicknesses effect on film surface topography parameters such as roughness, particle size and Root mean square of grains are calculated. Atomic Force Microscopy (AFM) confirm that the distribution grains size appears nanostructure and homogeneous in all films. RMS increases from 1.54 nm to 3.98 nm with thicknesses 500 nm. The surface roughness increases from 1.33 nm to 3.30 nm. Transmittance was detecting to be atop 80% in visible region. The bandgap energy increased from 2.83 eV to 3.75 eV with thickness elevation.

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1. Introduction

ZnO is has a unique characterization such as n – type semiconductor, high transparency and electron mobility, high thermal conductivity, wide bandgap energy of 3.4 eV [1-3], and excitation energy about 60 meV [4]. ZnO was entered wide applications such as gas sensing [5], ultraviolet laser emission [6], photodetectors [7], DSSCs [8], LEDs [9] and piezoelectric transducer [10]. Beyond the plethora of potential uses, the inexpensive cost of zinc oxide. ZnO is resistant to oxygen and moisture, has very good optical transparency, and has a flexible host crystal lattice able to accept a variety of dopant substitutions. ZnO Nanostructure thin films can be deposited by many methods. includes spin coating [11], chemical vapor deposition [12], SPM [13-16], dip coating [17], hydrothermal growth [18], electrodeposition [19]. RF sputtering [20], atomic layer deposition [21], thermal vacuum evaporation [22]. SPM is applied in producing (thin, multilayer, thick, and porous) film, which can produce large-area films with good optical properties and crystalline at a low cost. [23]. This work tends to prepare ZnO films with various thickness. different thickness thin films have been used to develop several thin films while maintaining stable substrate temperatures.

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2. Experimental

Thin films of ZnO were prepared on to glass substrate kept at 400°C with different thicknesses (300, 400 and 500) nm utilizing chemical spray pyrolysis method. 0.1 M $ZnCl_2$ was resolve in re-distilled water to get ZnO. A clear solution was fetched by adding few drops of HCl. The preparation conditions, which was applied were: space among spout and nozzle was 30 cm, spraying period 8 s, waiting for 60 s to prevent cooling, spraying rate was 5ml/min, N_2 was applied as a transporter gas. XRD is applied to locate film structure, whereas AFM was utilized to determine film morphology. UV-Visible spectrophotometer was used to specified absorption spectra in the 300-900 nm wavelength range.

3. Results and discussions

XRD styles are offered in Fig.1. These films exhibit polycrystalline with hexagonal wurtzite structure. This result was fit well compared with ICDD card no.35-1451. Maximum peak was noticed along (100), another planes were (101) and (102). The deposited films have a grain size of 26.61 and 38.02 nm for various thicknesses, that was obtained by Debye-Scherrer's equation 1 and assure film nanostructure, these films are revealed that grain size increases with increasing of thicknesses, similar behavior was reported by [24, 25]. Their values are summarized in Table 1. Grain size (D) was estimated by equation [26-28]:

$$D = \frac{0.9\,\lambda}{\beta cos\theta} \tag{1}$$

where β is FWHM), λ = 1.54Å and θ is Bragg's angle. The dislocation density (δ), is obtained via equation [29-31]:

$$\delta = \frac{1}{D^2} \tag{2}$$

Film microstrain (ε) is obtained via equation [32-34]:

$$\varepsilon = \frac{\beta cos\theta}{4} \tag{3}$$

Fig. 2 displays That ε and δ show slow decreasing in their values. The reduction in (ε) and (δ) might be due to locomotion of interstitial Zn atoms, that leads to a reduce in lattice defects [35-37].

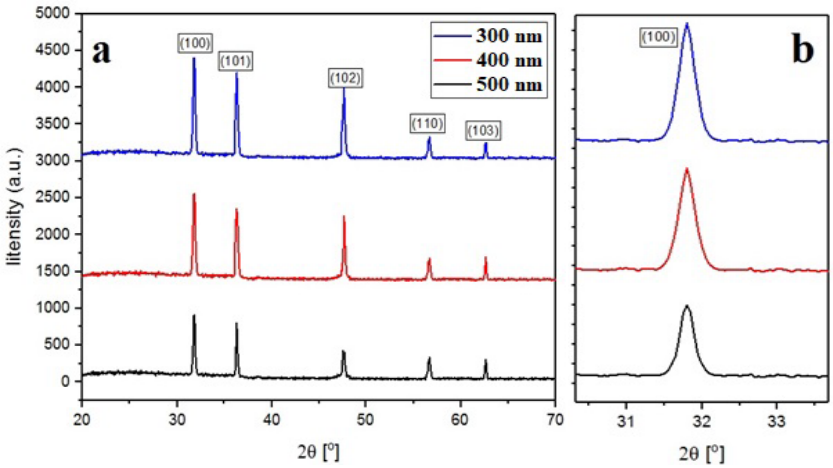

Fig. 1. XRD styles of the intended films.

Table 1. Structural parameters of the deposited films.

Samples nm	(hkl) Plane	2Θ (°)	FWHM (°)	(D) (nm)	ε (Line^{-2}·m^{-1}) ×10^{-5}	(δ) (Line. m^{-2}) ×10^{15}	a (Å)	
							Standard	Calculated
300	(100)	31.79	0.3	26.61	1.41	1.26	3.249	3.250
400	(100)	31.79	0.27	29.57	1.14	1.13	3.249	3.246
500	(100)	31.79	0.21	38.02	6.92	8.80	3.249	3.241

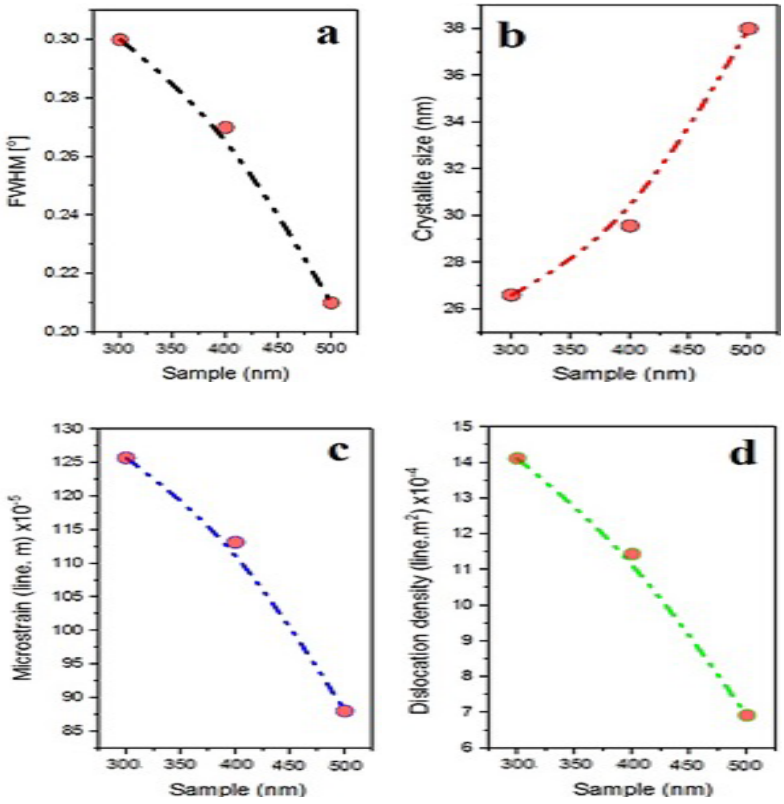

Fig. 2. Structure parameters of grown films.

4. Topography surface analysis

Fig.2 (a_1, a_2, a_3) shows AFM image of two and three dimensional images of the deposited films. Different film surface topography was observed. The roughness of these films increases from 1.33nm for 300nm to 3.3 nm for 500 nm as displayed in Fig. 2. At high thickness small assemblage grains were noticed lead to fashioning of large grains and increasing in surface roughness [38-39]. The maximum RMS value was found 3.98 for the 500 nm, which exhibited the larger grains size. As displayed in Table 2.

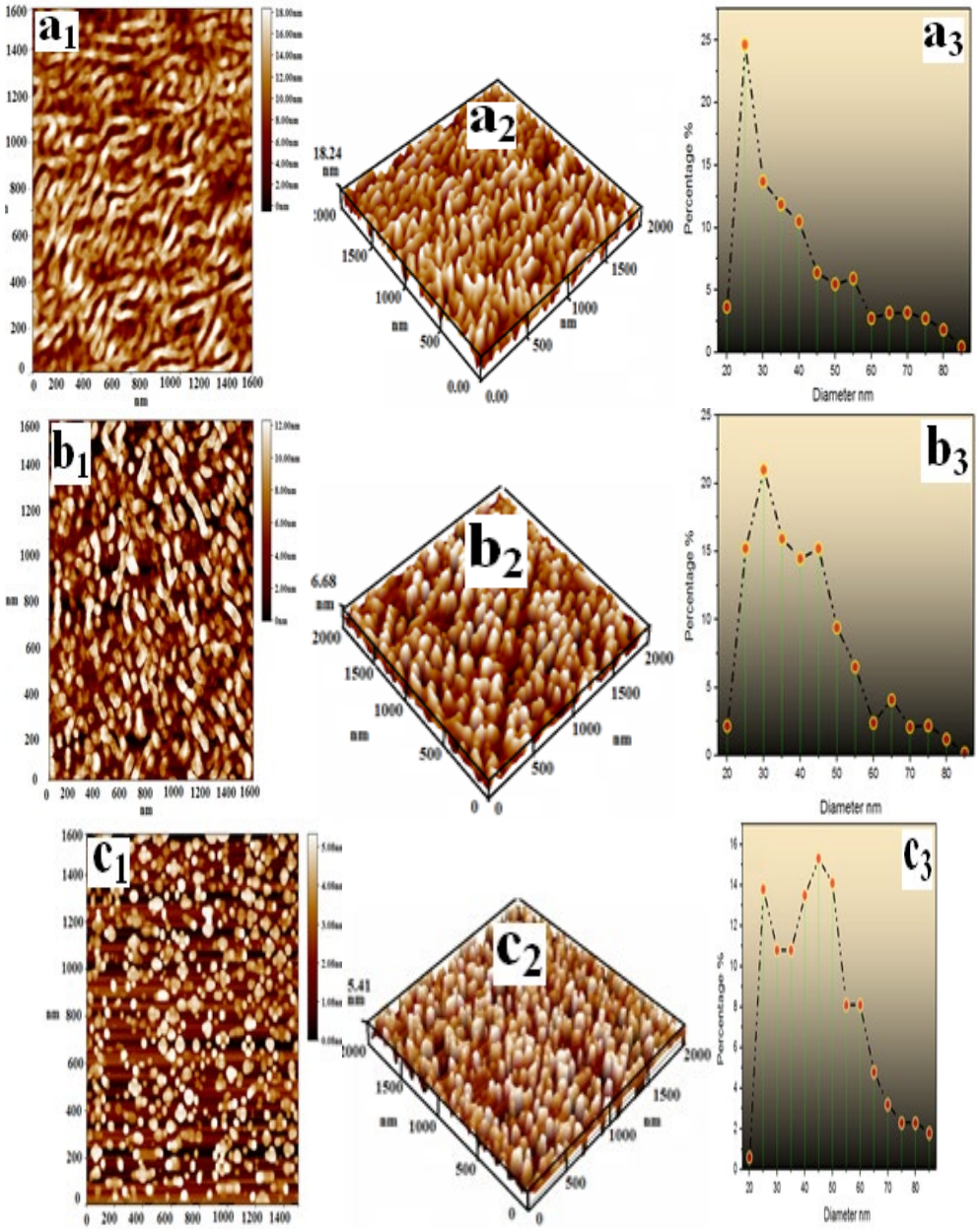

Fig. 3. AFM Images of deposited films.

Table 2. Surface morphology of ZnO films at different thickness.

Samples nm	Average Particle size nm	Average roughness nm	RMS nm
300	25	1.33	1.54
400	30	2.02	2.36
500	60	3.30	3.98

5. Optical properties analysis

The Transmission spectra of ZnO film is displayed in Figure 4. These films are exhibit transmittance above 80% in visible range. The transmittance was decrease with the increase in thickness. It can be explained by high surface scattering [40-43]. The value of bandgap E_g is evaluated the equation below [43-45]:

$$(\alpha h\nu) = A\left(h\nu - E_g\right)^{\frac{1}{2}} \tag{4}$$

Where A and $h\nu$ are constant and photon energy respectively. Figure 5 depicts the relation of $(\alpha h\nu)^2$ vs. $h\nu$ form which we can estimated E_g . The bandgap value of ZnO film for different thickness is 3.75, 3.65 and 2.83 eV. This could be attributed to the improvement in crystal order. [46, 47]. The absorption coefficient α is estimated by Beer-Lambert's law [48-50]:

$$\alpha = 2.303\frac{A}{d} = \frac{1}{d}\ln\left(\frac{1}{T}\right) \tag{5}$$

Fig.6 shows that α was decrease with thicknesses. This decrement might be due to increase in free carrier density [51-53]. The films deposited at 400 nm produced the highest α of ~7 x10^4cm^{-1} while at 500nm produced the lowest of ~1 x 10^4cm^{-1}. This result is very important in solar cells because the conversion efficiency depend on α, which influences in the solar conversion [54,55].

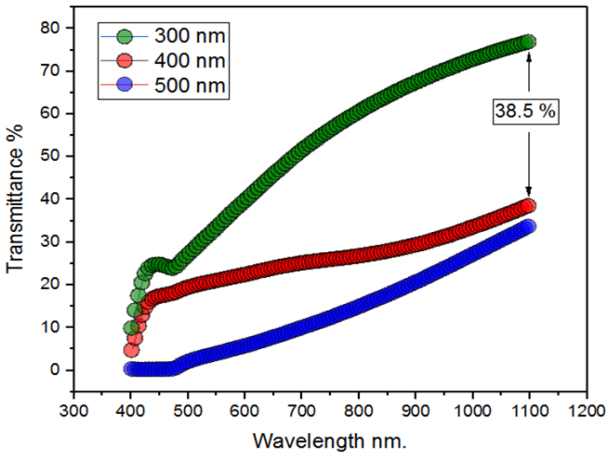

Fig. 4. Transmittance with wavelength of the deposited films.

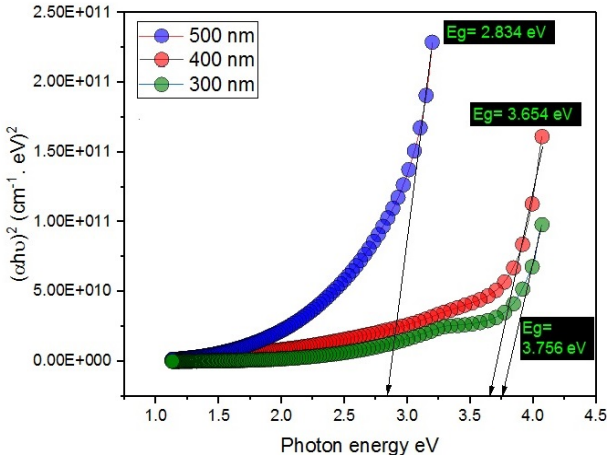

Fig. 5. $(\alpha h\upsilon)^2$ versus $(h)\upsilon$ of the deposited films.

6. Conclusion

Different thicknesses of ZnO were prepared by SPM. XRD assure that deposited films were polycrystalline with a hexagonal wurtzite structure. AFM surface topography of ZnO films revealed that roughness increases from about 1.33nm for 300nm to 3.3 nm for 500nm. It is found that these films have transmittance of above 80% in visible range. The bandgap reduces from 3.75, 3.65 and 2.83 eV, with the increase of thickness (300, 400 and 500) nm. All the films produced high α of the order of ($x10^4 cm^{-1}$).

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