



THE ROLE OF THICKNESS ON THE STRUCTURAL AND ELECTRICAL PROPERTIES OF DC MAGNETRON SPUTTERED NANO ZNO THIN FILMS

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Received 03-05-2012, revised 15-06-2012, online 26-06-2012

ABSTRACT

The objective of this research is investigating the effect of the thickness on the structural and electrical properties for nano ZnO thin films prepared by d.c magnetron sputtering on glass substrates. The deposition parameters such as power, target to-substrate spacing, substrate temperature and sputtering gas composition, self-heating of the substrate in plasma during film deposition were investigated. Thicknesses (t) of nanoZnO thin films are altered by varying the deposition time from 45 min to 90 min (t=50,100,150 and 200 nm) and was determined using optical method. Crystal structure were investigated by mean of a X-ray diffraction (XRD), it was found that all the films have peak located at $2\theta \approx 34.4^\circ$ with hkl (002), in addition there were another peaks appear at thickness 100 nm, 150 nm and 200 nm. The crystalline was increased with increasing (t). The grain size increased from 9.462 nm to 11.8522 nm as the film thickness increased from 50 to 200nm. General observations indicate that microstructure parameters, such as grain size, depend sensitively on the exact nature of the deposition conditions.

The d.c conductivity (σ) for nano ZnO thin films has been studied as a function of thicknesses. From all samples we can be noticed that the resistivity (ρ) increased with increasing the temperature and film thickness, while the activation energies decreased. The carrier concentration and mobility have been studied.

Key Word: Nano ZnO thin Films, Structural properties, Electrical properties, dc magnetron sputter.

I. INTRODUCTION

Zinc oxide (ZnO) is II–VI compound n-type semiconductor with a hexagonal wurtzite structure [1]. Recently, Zinc oxide is a versatile material with a wide direct band gap of 3.37 eV at room temperature [2]. ZnO has been extensively studied for various applications such as varistors transducers, transparent conducting electrodes, sensors, catalysts, UV light-emitting devices, transparent high power electronics, optical waveguides and solar cells [2-7]. ZnO also has a number of advantages over GaN the wide-band gap semiconductor currently utilized in the short-wavelength optoelectronics industry. Some of these advantages include a large exciton binding

energy (~60 meV) [8,9] a higher radiation hardness, simplified processing due to amenability to conventional chemical wet etching and the availability of large area substrates at relatively low material costs. Non-toxicity and relatively low deposition temperature [10, 11] .Researchers have found that the electrical properties strongly depend on the thickness of ZnO films. In this paper, we investigate the effect of thickness on the structural and electrical properties of ZnO thin films prepared by d.c magnetron sputtering which is an easy and economic process for the growth of polycrystalline ZnO thin films.

II. EXPERIMENTAL PROCEDURE

Nano ZnO thin film was fabricated by using d.c magnetron sputtering technique with inert gas like argon as the medium for glow discharge under different thickness (50,100,150 and 200 nm). Thicknesses (t) of ZnO thin films are altered by varying the deposition time from 45 min to 90 min and were determined using optical method.

$$t = \lambda_l \Delta x / 2x \quad (1)$$

where x is the fringes width, Δx is the fringes shift, and λ_l is laser wavelength. The structure of the films was characterized by X-ray diffraction (XRD) with Cu $K\alpha$ radiation and X-ray wavelength (λ) is 0.15406 nm. The average grain size (D_{XRD}) in the c-axis orientation estimated using the Debye-Scherrer relation [12]:

$$D_{XRD} = 0.9\lambda / B \cos \theta \quad (2)$$

Where θ is the Bragg diffraction angle and B is the full width at half maximum (FWHM) of the diffraction peak. The calculating of inter planer distance d for all set of films using the Bragg equation [13]:

$$d = n\lambda / 2\sin\theta \quad (3)$$

where n is the order of diffraction and λ is x-ray wavelength . Ohmic contacts for the prepared films were produced by evaporating Al electrodes of about 300 nm thickness, by means of thermal evaporation methods, using Edward coating unit under vacuum (10^{-5} mbar).

The d.c conductivity (σ) has been studied using the electrical circuit which was consisted of oven and Keithely electrometer.

D.c conductivity (σ) of samples obtained using equation [10] as $\sigma = L / R \cdot t$. where L is distance separated the electrodes, R is resistance of film. Then the activation energy (E_a) can be calculated from the equation [14]:

$$\sigma = \sigma_0 \exp (E_a / k_B T) \quad (4)$$

where T is absolute temperature, σ_0 is minimum metallic conductivity and k_B is Boltzmann constant.

From Hall Effect we can know the type of prepared films and carrier concentration and mobility.

III. RESULT AND DISCUSSION

Fig. (1) shows the XRD for ZnO thin films deposited on glass substrate at different thicknesses (50, 100, 150 and 200 nm). The patterns show that all the films have a peak located at $2\theta \approx 34.4^\circ$ with hkl (002), in addition another peaks are appeared at thickness 100, 150 and 200 nm located at $2\theta=31.80^\circ$, 36.30° and 56.58° with hkl{(100), (101), and (110)} respectively. All set of films reveal only an intense diffraction peak around 34° , which corresponds to (002) of the wurtzite phase of ZnO. This indicates that the growth of all films is highly oriented along c-axis [11]. Table (1) shows all parameters deduce from XRD pattern, the peaks observed and standard peaks from JSPDS [15], grain size (D_{XRD}) and its intensities.

Our result declared a good coincidence with the reference data and illustrate that the film have a good crystalline with hexagonal structure and the crystallinity increasing with increasing the thickness.

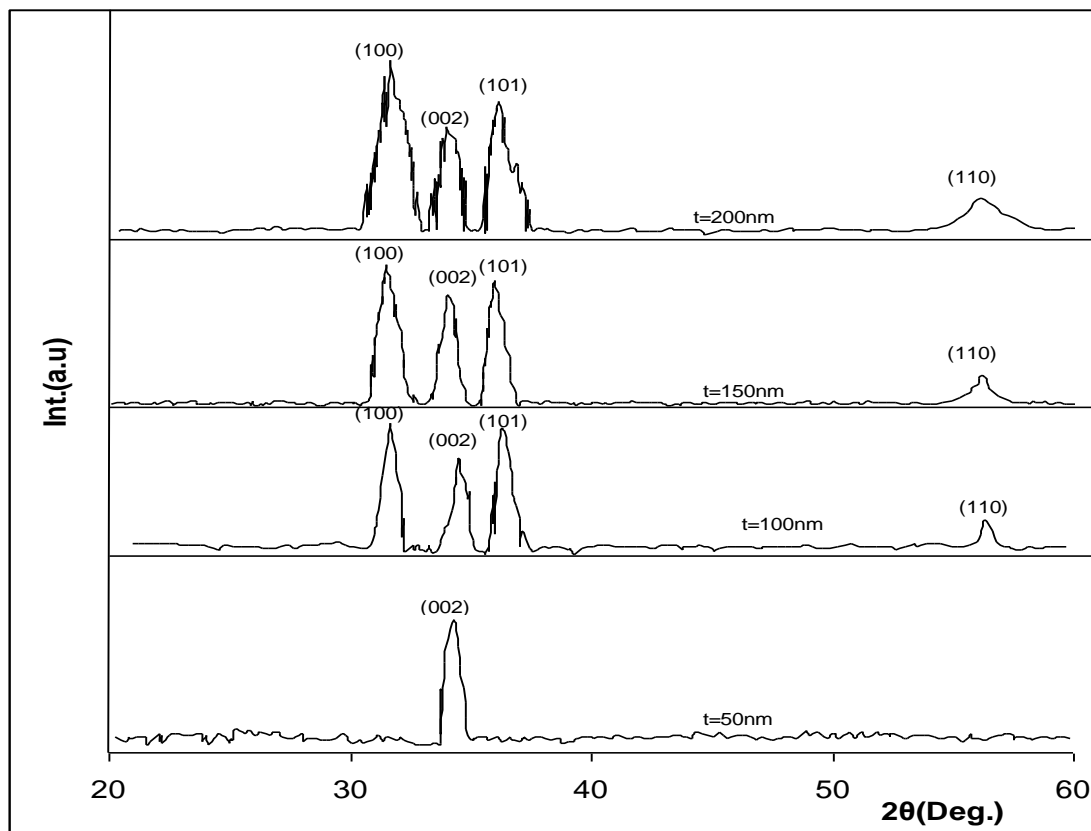


Figure 1: XRD pattern for ZnO thin films at different thicknesses.

Also we can see from Table (1) the grain size of the prepared films at (002) plane is increased from 9.462 nm to 11.852 nm when the film thickness increased from 50 to 200 nm respectively. Therefore, it is observed that the increased thickness provided a better crystallization for the films.

Table 1: XRD parameter (experimental and standard): 2θ , d , intensity and grain size for prepared ZnO thin films as a function of thickness

Thickness (nm)	2θ (exp)(deg) at peak	d_{hkl} (Exp.) (\AA°)	Int.%	2θ Stan. (deg)	d_{hkl} (Stan.) (\AA°) [15]	Int.%	(hkl)	D_{XRD} (nm)
50	34.363	2.607	60	34.241	2.603	44	002	9.462
100	36.253	2.475	93	36.252	2.475	100	101	
	31.821	2.809	100	31.769	2.814	57	100	
	34.406	2.604	60	34.241	2.603	44	002	10.904
	56.160	1.636	10	56.602	1.911	32	110	
150	36.265	2.475	61	36.252	2.475	100	101	
	31.793	2.812	82	31.769	2.814	57	100	
	34.405	2.604	100	34.241	2.603	44	002	11.357
	56.200	1.635	10	56.602	1.911	32	110	
200	36.252	2.475	100	36.252	2.475	100	101	
	31.816	2.810	98	31.769	2.814	57	100	
	34.414	2.603	50	34.241	2.603	44	002	11.852
	56.250	1.634	10	56.602	1.911	32	110	

The d.c. conductivity for ZnO films has been studied as a function of temperature within the range of (303-543 K) at different thicknesses (50, 100, 150 and 200nm) as shown in Fig. (2). This figure declared that all prepared ZnO thin films have two stages of conductivity throughout the heating temperature range. The first activation energy (E_{a1}) occurs at higher temperature and this activation energy is due to conduction of the carrier excited into the extended states beyond the mobility edge, while the second activation energy (E_{a2}) occurs at low temperature and the conduction mechanism of this stage is due to carriers transport to localized states near the valence and conduction bands [14].

From all the samples we can notice that the d.c. conductivity at room temperature (σ_{RT}) increases with increasing the temperature in other word the variation of the resistivity for the layers according to the thickness the resistivity decreases with the thickness. The increase in the conductivity can be attributed to the increase in the grain size and the high carrier concentration because of the interstitial zinc atom and oxygen vacancy which act as donor. The increase in the grain size caused to decreases in grain boundaries. This result is in a good agreement with the literature [10].

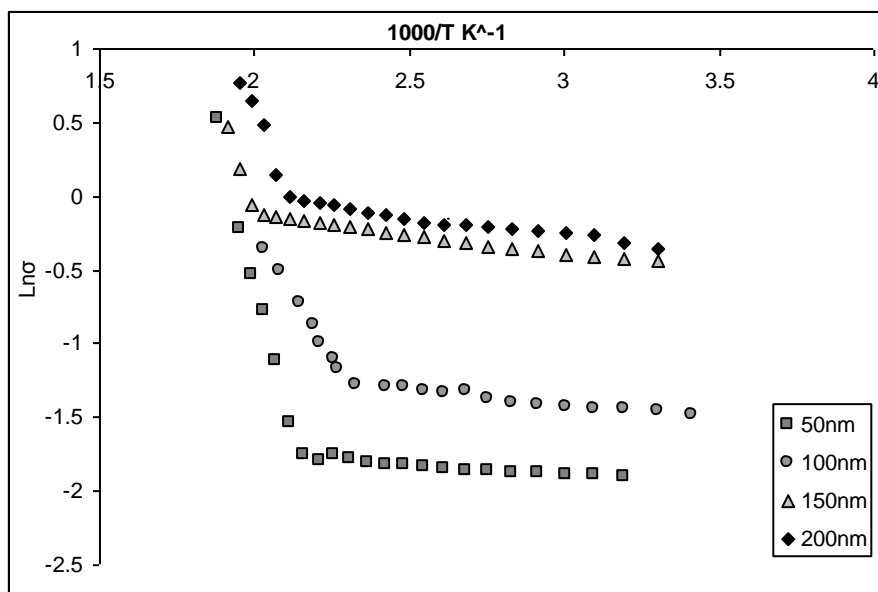


Figure 2:The variation of $\ln\sigma$ with $1000/T$ for ZnO thin films at different thicknesses.

Table (2) shows the E_{a1} , E_{a2} and temperature range for different film thickness. We can notice from this Table the decreasing of activation energies with the increasing of film thickness.

Table 2: Values of $\sigma_{(RT)}$, E_{a1} and E_{a2} for ZnO thin films at different film thickness.

Thickness(nm)	$\sigma_{(RT)}$ ($\Omega.cm$) ⁻¹	E_{a1} (eV)	Temp range (K)	E_{a2} (eV)	Temp range (K)
50	7.10×10^{-3}	0.97	473-543	0.106	293-473
100	2.20×10^{-3}	0.700	433-543	0.054	293-433
150	3.50×10^{-2}	0.685	500-543	0.038	293-500
200	6.41×10^{-1}	0.600	476-543	0.0225	293-476

From Hall measurement we can observe that all samples are n-type. The carrier mobility and concentration of ZnO thin films monotonically increased with increasing the film thickness. With increasing thickness for the thinner films, the increase carrier mobility and concentration is attributed to the improved crystallinity and increased crystallite sizes that weakens inter-crystallite boundary scattering and increases carrier lifetime [13]. The dependence of the carrier mobility and concentration on the thickness of ZnO thin films deposited with self heated from 20 to 125 °C substrate temperatures was shown in Table (3). It can be seen that the carrier concentration and mobility increased about two orders of magnitude respectively. Figure (3) showed the variation of carrier concentration (N_H) and mobility (μ) with thickness of the films.

The resistivity (ρ) is proportional to the reciprocal of the product of carrier concentration (N_H) and mobility (μ) ($\rho=1/N_H e \mu$)[12]. This lower resistivity of ZnO film at average of substrate temperature 373K was caused by the higher product of carrier concentration (N_H) and mobility (μ) Figure (4) showed the variation of resistivity (ρ) and mobility (μ) with thickness of the film .These results appear agreement with the researches [16].

Table 3: The dependence of the R_H , N_H , μ , and resistivity on the thickness for ZnO thin films.

Thickness (nm)	R_H	$N_H(\text{cm}^{-3})$	$\mu (\text{cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1})$	$\rho(\Omega \cdot \text{cm})$
50	-3.70×10^5	1.70×10^{13}	1.44×10^2	2553.1
100	-3.67×10^4	1.76×10^{14}	3.3×10^2	111.408
150	-2.56×10^4	2.44×10^{14}	1.35×10^3	18.973
200	-1.23×10^3	5.03×10^{15}	2.50×10^3	2.012

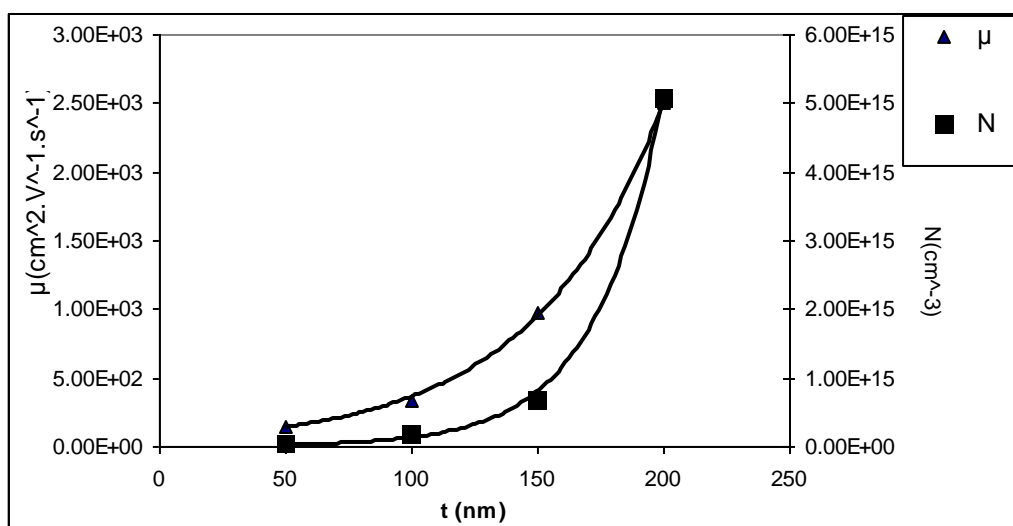


Figure 3: The variation of carrier concentration and mobility with thickness for ZnO thin film.

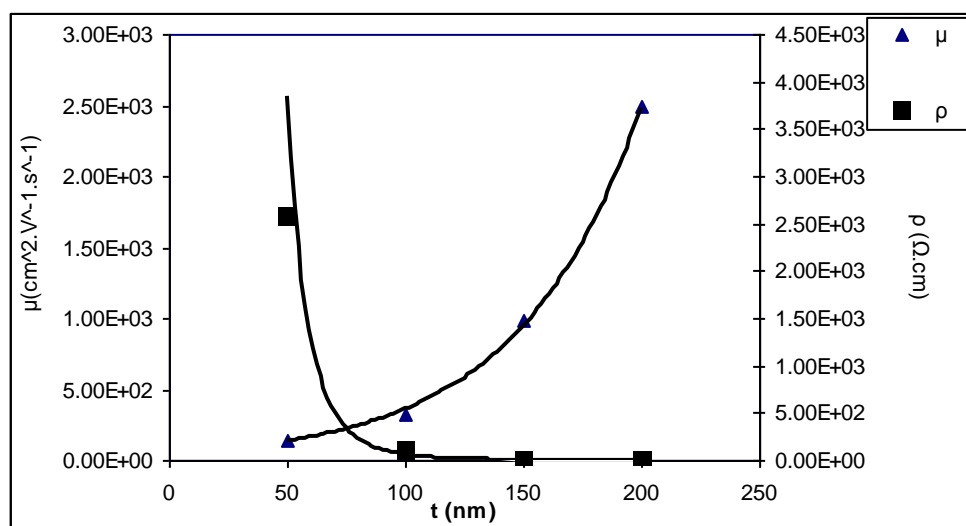


Figure 4: The variation of resistivity and mobility with thickness for ZnO thin film.

IV. CONCLUSIONS

Conductive and transparent nano ZnO thin films were deposited successfully by Zn target using d.c magnetron sputtered method, and the corresponding characteristics of the films as a function of thickness were reported:

- Polycrystalline ZnO thin films have demonstrated three important planes of hexagonal structure (100,200 and 101) for various thicknesses.
- The grain size increased with increasing the film thickness.
- The film conductivity increased with increasing thickness, while the two activation energies decreased.
- The type of all prepared thin films are n-type.
- The carrier concentration and mobility of all prepared samples increased with increasing thickness.

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