Synthesis and Characterizations of ZnO thin films deposited on aluminum metal substrate by sol-gel spin coating technique

Ali Shaitir [a], A.G.E. Sutjipto [b*], Y. Shian [c]
Faculty of Industrial Sciences & Technology
University Malaysia Pahang
Lebuhraya Tun Razak, 23600 Kuantan Pahang, Malaysia
[a] alishaiter@yahoo.com, [b] agusgeter@ump.edu.my

Abstract— In this work, zinc oxide (ZnO) thin films were synthesized on aluminum substrate using sol-gel spin coating technique. The effect of changing in the annealing temperature (from 300 ◦C to 500 ◦C) and the rates of spin (from 1000rpm to 2000rpm) on the structural, optical and piezoelectric properties of the produced films are studied. Zinc acetate dehydrate, absolute ethanol and diethanolamine were used to act as sol gel precursor. Sol gel was coated on glass slide which wrapped by aluminum. The thin film was formed after preheating and annealing. The thin film was characterized by X-ray diffraction (XRD), Photoluminescence Spectroscopy (PL) and Ultraviolet-visible Spectroscopy (UV-Vis). X-ray analysis showed that thin films were preferentially diffracted around 65° which corresponding to (1 1 2) diffraction phase. From the PL results, there was only film with spin speed of 2000 rpm and annealing temperature of 300°C had slightly left wavelength which was 380 nm. Annealing temperature would affect only the intensity of PL wavelength. From the results of UV-Vis, it was observed that when the spin speed was increased at same annealing temperature, the band gap was decreased. When the annealing temperature was increased at same spin speed, the band gap was decreased. This work provides a low cost and environment friendly material for Vibration sensing applications.

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Keywords— Zinc oxide thin films; Sol-gel spin coating; Annealing temperature; Structural properties; Optical properties; Piezoelectric properties

1. INTRODUCTION

Zinc oxide is a non toxic, abundant, low cost and wide direct band-gap (3.2–3.4 eV) II–VI compound semiconductor [1–3]. This leaves it within the visible region having high optical transmission, suitable for solar spectrum [1]. Therefore, it has vast applications in optoelectronics device because of its structural, electrical and optical properties [4–9]. Due to these promising characteristics, thin films of ZnO have showed the considerable attention in the literature [1,10–12]. It is proposed that Multilayered ZnO thin films can increased the electrical properties of films [2]. Optimized multilayered ZnO films can be designed to have low resistivity and high solar transparencies in the visible region that are needed for the applications in solar cells and light emitting diodes [13].

The low cost and small time require for the synthesis of ZnO thin films for industrial requirements is always a need. Multilayered thin films of ZnO can be deposited by several methods such as magnetron sputtering [14], pulsed laser deposition [15] and spray pyrolysis [16]. Among them sol-gel technique is simple, low cost and large area deposition technique.

The review of literature showed that the properties of stacked ZnO thin films layers, deposited by sol-gel technique, which is cost effective and easy deposition technique, has not been studied yet. Current study will facilitate to fabricate optoelectronic devices with affordable cost. In present work, the structural, electrical and optical properties of multilayers of ZnO thin films deposited by sol-gel spin coating has been discussed [22-24].

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2. METHODOLOGY

The stacked layers of ZnO thin films were prepared by using solgel method. The ZnO solution was prepared by dissolving 0.88 g of ‘‘Zinc acetate dehydrates” in the 20 ml of ‘‘2-methoxyethanol”. Then this solution was stirred with the help of magnetic stirrer at 60 º C for 30 min. After that the ‘‘Mono ethanol amine” as a stabilizer was added in this solution and stirred the solution for 90 min at the temperature of 60 º C. The clear homogeneous solution of ZnO was obtained by aging this solution for 24 h.

The solution is now ready to prepared films. Now the aluminum substrate is fixed on spin coating system and its disc speed is changing (from 1000rpm to 2000rpm). The rotation time is 30s per sample. The drops of sol-gel solution are dropped on the aluminum substrate as per requirement of layers. After 30 s, the aluminum substrate is removed from spin coating disc and placed on the hot plate for almost 10 min to dry at 100 º C. The experiment is repeated to make thin films with multilayer.

The thin films are put in Furnace with changing in the annealing temperature (from 300 ˚C to 500 ˚C). Then the furnace is switched off and each sample is left to cool down until it reaches room temperature. Then samples are taken from the furnace and now they are ready to analyze.

3. RESULTS AND DISCUSSION

A. XRD results

From Figure 1, the angle of highest peak was ~45°. The lattice parameters of films were a = 3.4 Å and c = 5.1 Å. These values were similar to the values reported for good quality piezoelectric ZnO thin films [17]. For the plane of ZnO, it could be found that all the films were preferentially oriented in the (1 1 2) direction at ~65°. The presence of the (0 0 2) peak in some films, indicates that the as-deposited ZnO thin films have strong c-axis orientation perpendicular to the substrate. Higher degree of c-axis orientation and (0 0 2) preferred crystalline structure improve the piezoelectric properties of the ZnO thin film [18]. The presence of (0 0 2) diffraction phase was due to the precursor solution used was ZnAc which promotes the growth of crystal in (0 0 2) direction [19]. Some of the films could not find the peak of (0 0 2) may due to destructive interference happened.

Table 1 shows the average crystallite size of ZnO films with different spin speed and annealing temperature. When the annealing temperature was increased, the crystallite size was increased. The crystallite size was decreased from 0.5832 nm to 0.4955 nm and to 0.4512 nm. It had shown that temperature could affect material crystal sizes.

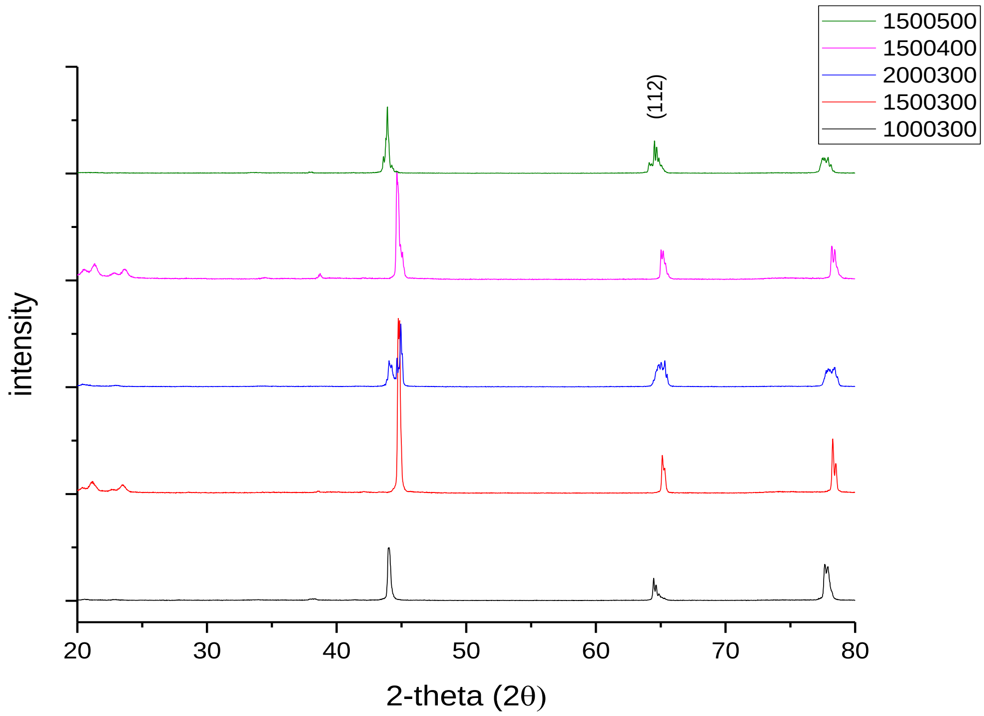

Figure 1: XRD pattern of ZnO thin films.

Table 1: (h k l) plane, d-spacing and crystalline size for the ZnO thin films with different spin speed and annealing temperature.

Sample	Spin Speed (rpm)	Anneal Temperature (°C)	2θ (°)	(h k l)	d-spacing	FWHM	Average Crystalline Size, nm
A	1000	300	43.9918 64.4470	(1 0 1) (1 1 2)	2.0566 1.44570	0.181 0.174	0.8405
B	1500	300	65.1421 74.0400	(1 1 2) (0 0 4)	1.43083 1.27900	0.151 2.260	0.5832
C	2000	300	34.3700 65.0830 78.3940	(0 0 2) (1 1 2) (1 0 4)	2.60700 1.43199 1.21882	1.100 0.260 0.256	0.4878
D	1500	400	34.4700 65.0300 74.1000	(0 0 2) (1 1 2) (0 0 4)	2.60000 1.43303 1.27800	0.560 0.142 2.500	0.4955
E	1500	500	33.6100 64.8820 75.5000	(0 0 2) (1 1 2) (2 0 2)	2.66400 1.43594 1.25900	0.900 0.143 4.000	0.4512

B. PL results

PL spectra of ZnO films were shown in Figure 2. The emission spectrum of ZnO thin film showed the peak was at narrow band ~380 nm. It was corresponding to band edge emission. It was due to free exciton emission. It has ultraviolet region. The PL spectrum also showed a wide broad peak of green emission at ~500 nm. The green emission was due to the existence of intrinsic defects, which are zinc vacancies and oxygen interstitial [20]. It could be observed that when the annealing temperature was increased, the wavelength intensity was increased. This was because of the oxygen and zinc atoms in interstitial site moved to lattice sites. It was also due to surface grain area increases [21]. From Figure 2, it can be seen that there was insignificant change in wavelength; this is because the wavelength shift only caused by composition of precursor but not annealing temperature and spin speed. Annealing temperature had only changed the intensity of wavelength [19].

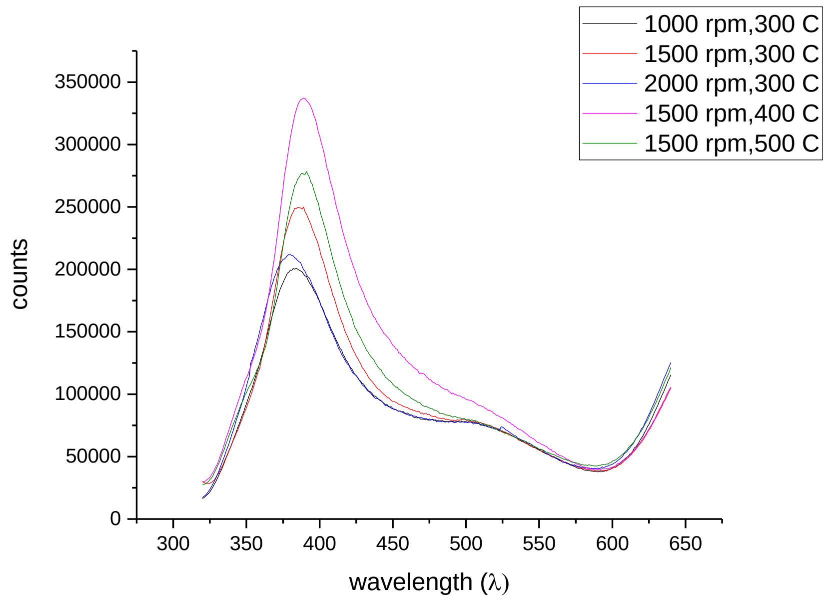

Figure 2: PL spectrum of ZnO thin film with different spin speed and annealing temperature.

C. UV-Vis Analysis

From Figure 3 to Figure 7 show the UV emission spectrum of ZnO with different spin speed and annealing temperature. Table 2 showed the energy band gap of each ZnO film with different spin speed and annealing temperature. It could be found that when increasing the annealing temperature, the energy band gap was decreased. The highest band gap (3.6109 eV) was belong to ZnO film with spin speed of 1000 rpm and annealing temperature of 300 °C.

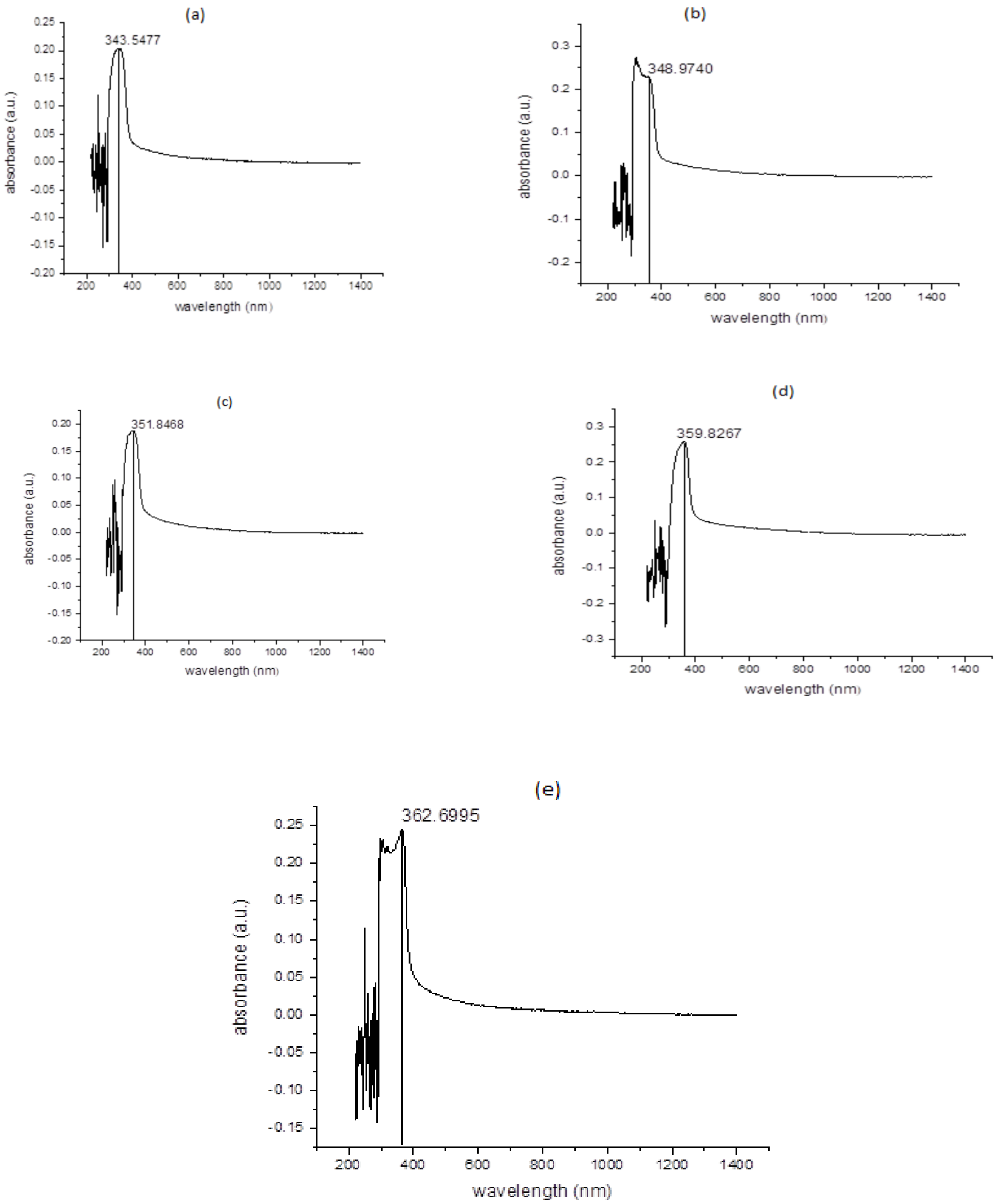

Figure 4: UV spectrum of ZnO film with (a-1000 rpm and 300 °C), (b-1500 rpm and 300 °C), (c- 2000 rpm and 300 °C), (d- 1500 rpm and 400 °C), and (e- 1500 rpm and 500 °C) respectively.

Table 2: Energy band gap of UV emission peak at different spin speed and annealing temperature of ZnO thin film.

Sample	Spin Speed (rpm)	Anneal Temperature (°C)	First Excitonic Peak (nm)	Energy band gap (eV)
A	1000	300	343.5477	3.6109
B	1500	300	348.9740	3.5547
C	2000	300	351.8468	3.5257
D	1500	400	359.8267	3.4475
E	1500	500	362.6995	3.4202

4. CONCLUSIONS

In summary, ZnO multilayer thin films were prepared by sol-gel method. XRD confirms the hexagonal wurtzite structure. PL results shows that, there was only film with spin speed of 2000 rpm and annealing temperature of 300°C had slightly left wavelength which was 380 nm. Results of UV-Vis shows that, the band gap was decreased when (the spin speed or the annealing temperature) were increased.

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