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# EFFECT OF SPUTTERING PRESSURE ON THE STRUCTURAL AND OPTICAL PROPERTIES OF ZnO FILMS DEPOSITED ON FLEXIBLE SUBSTRATE

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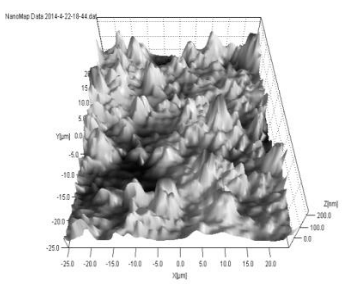
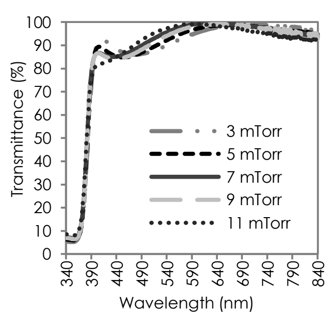
**Graphical abstract**






## Abstract

Zinc Oxide (ZnO) has get attention because of its excellent optical transmittance approximately ~80%. A systematic study has been made of the effect of sputtering pressure on the structural and optical properties of the deposited ZnO films. ZnO thin films were deposited on plastic substrate by RF powered magnetron sputtering using ZnO disk with 99.99% purity. The RF power of 100 W and argon gas flow from 10 - 30 sccm was used to perform the sputtering process. The argon sputtering pressure was varied between 3 mTorr to 11 mTorr and the growth time is constant at 25 minute. The deposited ZnO thin films are characterized structurally and optically. Structural analysis using X-Ray Diffraction (XRD) show that all the films exhibit a (002) preferential orientation and the estimated grain size measured was varied from 24.5 nm to 29.8 nm. Surface Profilometer was used to measure the thickness and RMS of the deposited films where the thickness decrease from 900 nm to 600 nm and the root mean square (RMS) roughness increase from 4.97 nm to 11.45 nm. The optical transparency value that achieved from UV-Vis Spectrometer is over 80% in the visible range and the band gap energy is within 3.27 eV-3.29 eV range.

*Keywords*: Zinc Oxide, flexible substrates, RF powered magnetron sputtering

## Abstrak

Zink Oksida telah mendapat perhatian kerana penghantaran optik yang baik dalam kira-kira ~ 80%. Satu kajian sistematik telah dibuat dari kesan tekanan pemercikan pada sifat-sifat struktur dan optik filem-filem ZnO yang didepositkan. Filem nipis ZnO telah didepositkan di atas substrat plastik dengan menggunakan RF berkuasa pemercikan magnetron menggunakan cakera ZnO dengan ketulenan 99.99%. Kuasa RF 100 W dan aliran gas argon dari 10-30 sccm telah digunakan untuk melaksanakan proses pemercikan. Tekanan argon pemercikan telah diubah antara 3 mTorr kepada 11 mTorr dan masa pertumbuhan adalah malar pada 25 minit. ZnO filem nipis yang didepositkan dicirikan secara struktur dan optik. Analisis struktur menggunakan belauan X-Ray menunjukkan bahawa semua filem menunjukkan orientasi utama (002) dan saiz anggaran butiran yang diukur berubah daripada 24.5 nm kepada 29.8 nm. Permukaan profilometer telah digunakan untuk mengukur ketebalan dan kekasaran filem yang dideposit di mana ketebalan berkurang daripada 900 nm ke 600 nm dan punca min kuasa dua kekasaran permukaan meningkat daripada 4.97 nm kepada 11.45 nm. Nilai ketelusan optik yang dicapai daripada UV-Vis Spektrometer adalah lebih 80% dalam julat yang boleh dilihat dan jurang jalur tenaga adalah dalam julat 3.27 eV-3.29 eV.

*Kata kunci*: Zink Oksida, substrat fleksibel, RF berkuasa pemercikan magnetron

## 1.0 INTRODUCTION

ZnO is one of the most promising transparent semiconductor oxide that can be apply in various electronics and optoelectronics devices for instance solar cells, light-emitting diodes and thin-film transistors [1]. ZnO had become an attractive choice as a semiconductor active layer in thin-film diode due its direct band gap of 3.37 eV, large exciton binding energy of 60 meV, high electrical conductivity, good ultraviolet absorption behavior, low cost, non-toxicity and easy fabrication [2].

There are several deposition techniques to fabricate ZnO thin films such as RF or DC magnetron sputtering [3, 4], sol gel process [5], spray pyrolysis [6], pulsed laser deposition [7], atomic layer deposition [8], electro-deposition [9] and chemical vapor deposition [10]. The fabrication of the ZnO on various substrates especially on flexible substrate required low deposition temperature. RF powered magnetron sputtering is considered as the suitable deposition technique that can be used. This technique allows the deposition at low temperature and widely been used for the fabrication because the advantages it has such as high deposition rate, low substrate temperature, good adhesion and dense layer formation [11, 12].

Recently, plastic or flexible substrates has many merits compared with glass substrates in many applications due to its lower cost, light weight, easily folded and are easy to carry [13, 14]. Some remarkable applications of the flexible substrate are flexible smart phone, digital book, gas sensor on lab coat and flexible organic light emitting diode. Other than that, plastic device that fabricated on thin plastic substrate are flexible and bendable to a large angle which mean the device can subjected to impact loading, rolled up for storage and mounted on curved surfaces without difficulty [15].

In this study, ZnO thin films were deposited on plastic substrates by using RF powered magnetron sputtering at room temperature. The influence of sputtering pressure on the structural and optical properties of ZnO thin films was investigated.

## 2.0 EXPERIMENTAL

ZnO films were deposited on plastic substrate using RF powered magnetron sputtering method. The target used is 3 inches diameter ZnO disk with 99.99 % purity. The substrates were ultrasonically cleaned for 5 minute in distilled water and then in ethanol and lastly in acetone twice each time. After that, the substrates were rinse with distilled water and dried it with nitrogen flow before deposition.

The sputtering processes are carried out with argon gas that flow from 10 to 30 sccm and the sputtering power adjusted to maintain at 100 W. The substrate rotation and deposition time were 5 rpm and 25 minute respectively. The sputtering was carried out under room temperature and the sputtering pressure was varied from 3 mTorr to 11 mTorr. The film thickness was measured using a NanoMap LS500 Profilometer. The structural and crystalline properties of the thin films were characterized using X-Ray Diffraction Philips Expert Pro. The optical properties of the ZnO films were analysed by using UV-Vis Spectrometer Lambda EZ210.

## 3.0 RESULTS AND DISCUSSION

Figure 1 shows the dependence of the growth rate and thickness of ZnO thin films on the sputtering pressure deposited on plastic substrates and the growth time is constant at 25 minute. The growth rate and thickness decreases as the sputtering pressure increases from 3 mTorr to 11 mTorr where the ZnO film thickness decreases from 900 nm to 600 nm. Similar result has been reported by Medina-Montes *et al.*, [1] and Hoon *et al.*, [16], where deposition rate and thickness of ZnO thin films decrease with an increase in the argon pressure. This happen due to the fact that lot of sputtered atoms reaching the surface of the substrate because the sputtered atoms experience larger mean free path with low collisions with argon plasma ions and atom which leading to the higher growth rate at low sputtering pressure. Meanwhile the sputtered atoms become scattered due to collision with each other and cause the growth rate decrease at high sputtering pressure [1, 16].

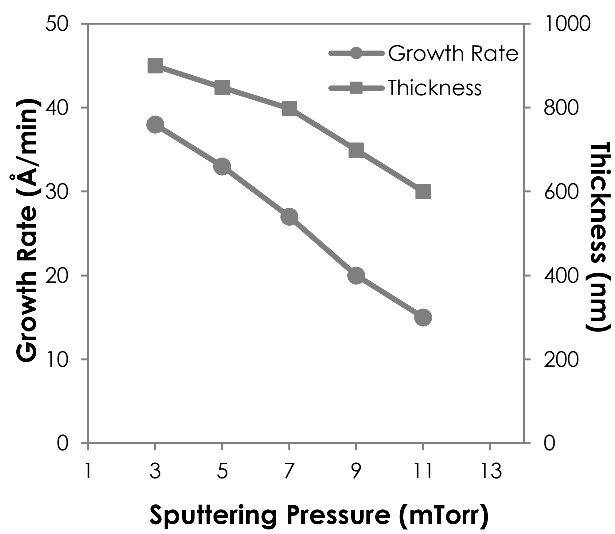



**Figure 1** Dependence of growth rate and thickness of ZnO thin films on the sputtering pressure deposited on plastic substrates and the growth time is 25 minute

Figure 2 shows the X-ray diffraction pattern of investigated ZnO films on plastic substrate with different sputtering pressure. From the XRD result, it revealed that all the films exhibit a (002) preferential orientation as the evidence of the high relative intensity of (002) diffraction peaks at about 34˚. As the sputtering pressure increased from 3 mTorr to 11 mTorr, the (002) diffraction peaks of ZnO films were shifted to the higher angle indicate that the lattice spacing between the (002) planes become smaller due to the increasing of uniform stress along the c-axis, agree with the result of *Hezam et al.*, [17].

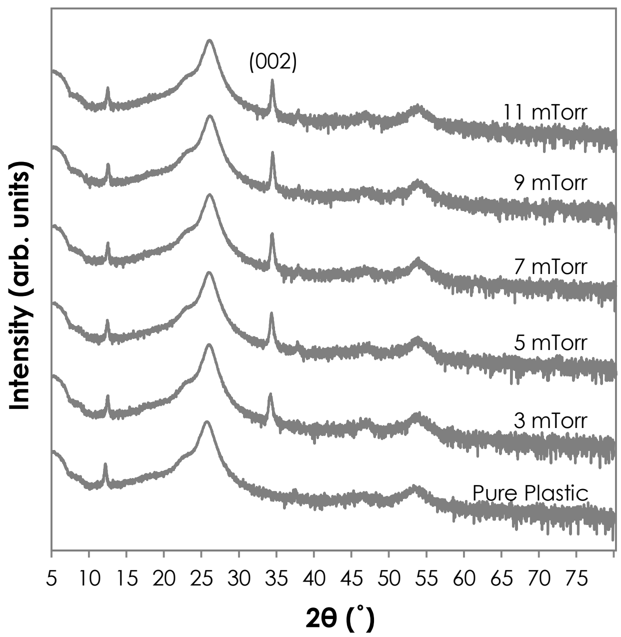



**Figure 2** X-ray diffraction patterns of investigated ZnO films deposited on plastic substrate at different sputtering pressure

Figure 3 shows the variation of full width at half maximum (FWHM) and estimated grain size of ZnO (002) diffraction peak at different sputtering pressure. The peak intensity became intense and the full width at half maximum becomes narrow where the value of FWHM decreases from 0.324˚ to 0.262˚ as the sputtering pressure increases. This indicates that the grain size of the films increase and the crystallinity of the films are improved with the increasing of sputtering pressure which similar with the finding of Lee [13] and Kim *et al.*, [18]. The XRD result can be used to calculate the estimated grain sizes of the films by using Scherrer's formula [12]:

$$D = 0.94\lambda/\beta cos\theta \qquad (1)$$

where X-ray wavelength, $\boldsymbol{\lambda}$ = 1.54056 Å, β is the FWHM of ZnO diffraction peak, and θ is the Bragg diffraction angle. The estimated grain size value increases from 24.5 nm to 29.8 nm as the sputtering pressure increase.

Table 1 shows the Profilometer images and RMS of ZnO thin films deposited at different sputtering pressure. The surface morphology revealed that the root mean square (RMS) roughness increases and the granular structure of the deposited films enhanced which consistent with the XRD result from Figure 3 where the grain size becomes larger with the increasing of sputtering pressure. The RMS roughness increases maybe due to the insufficient of energy for surface diffusion cause by the collisions with many atoms as the decreases of particles energy that arrives at the surface with the increasing sputtering pressure, which similar with earlier finding of Hoon *et al.*, [16] and Kim *et al.*, [12].

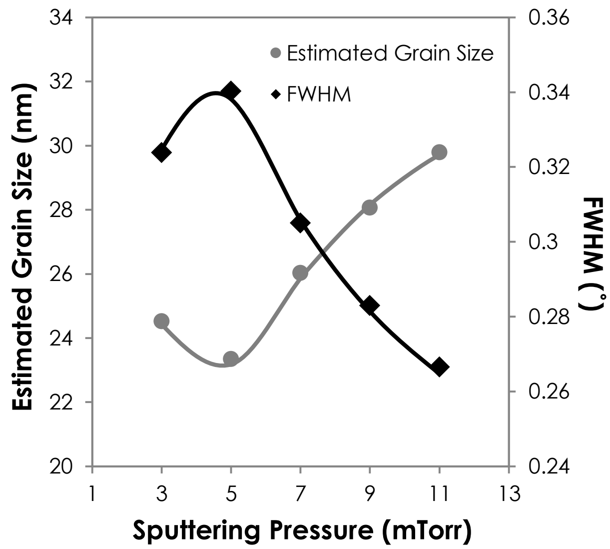



**Figure 3** The variation of full width at half maximum (FWHM) and estimated grain size of ZnO (002) diffraction peak at different sputtering pressure

Figure 4 shows the optical transmittance spectra of ZnO films deposited on plastic substrate at different sputtering pressure by RF powered magnetron sputtering at room temperature. High average transmittance of over 80 % was revealed for all the thin films which are in agreement with the result of Lee [13]. Based on the graph, argon sputtering pressure show significant different on the transmittance value where the optical properties improved as the sputtering pressure increase. The increasing of optical transmittance can be explain by the decreasing thickness and enhanced crystallinity of the ZnO thin films. A similar behavior was observed by Hoon *et al.*, [16]. The optical absorption coefficient (α) can be determined from the expression [18]:

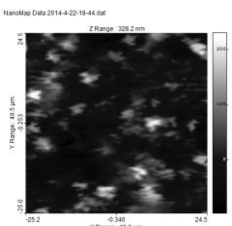
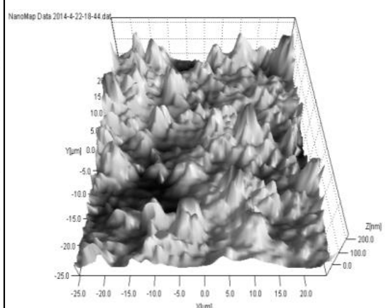
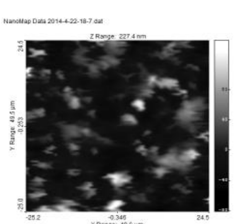
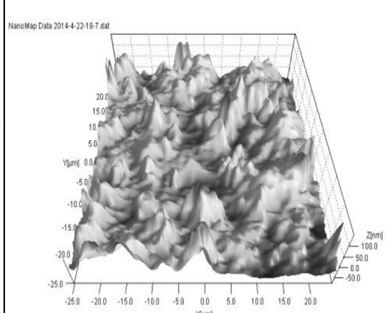
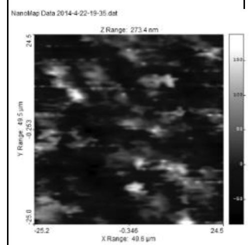
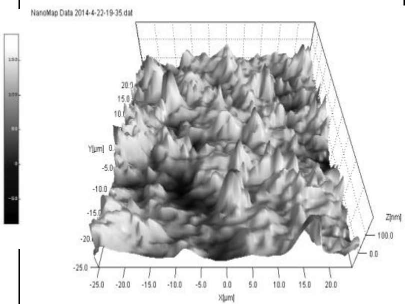
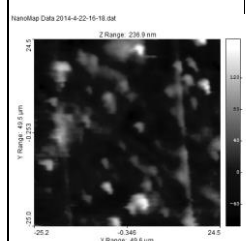
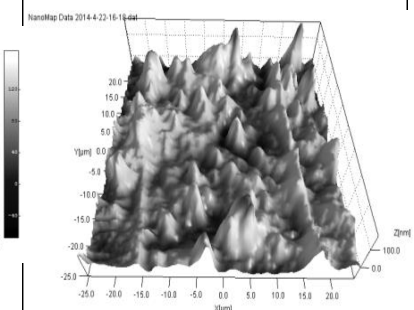
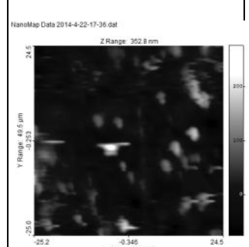
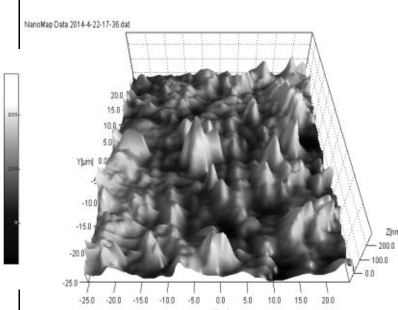
$$\alpha = (1/d)\ln T \qquad (2)$$

where $d$ is the film thickness and $T$ is the optical transmittance of the ZnO films. The optical band gap energy $(Eg)$ can be express using the following expression [18]:

$$(\alpha h\nu) = A(hv - Eg)^{1/2} \qquad (3)$$

where $Eg$ is the energy band gap, α is the absorption coefficient, $hv$ is the incident photon energy and $A$ is the edge width parameter. Figure 5 shows the plot of graph $(\alpha hv)^2$ versus photon energy different sputtering pressure. ZnO films on plastic substrate have direct band gap because of the dependence $\alpha hv = f(hv)$ is linear where the exponent $\mathrm{m} = 1/2$ . The values of optical band gap energy have been determined by the speculation of the straight line portion of this plot of graph to the photon energy axis. The estimated value of optical band gap was extrapolated at within 3.27 eV-3.29 eV range for all the deposited films and the values are similar with the previous study of Hezam *et al.,* [17]. As sputtering pressure increases, it is observed that the absorption coefficient decreased and the energy band gap value increases which agree well with the research of Lai *et al.,* [19]. The broadening of band gap with the increases of sputtering pressure may be due to the amorphous phase in ZnO films which consistent with the increases in the transmittance tails from Figure 5 [20].

**Table 1** 2D and 3D Profilometer images and RMS of ZnO thin films deposited at different sputtering pressure (a) 3 mTorr, (b) 5 mTorr, (c) 7 mTorr, (d) 9 mTorr and (e) 11 mTorr

| 2D | 3D |
|---|---|
| **(a) 3 mTorr** | |
|  | RMS: 4.97 nm  |
| **(b) 5 mTorr** | |
|  | RMS: 7.02 nm  |
| **(c) 7 mTorr** | |
|  | RMS: 8.12 nm  |
| **(d) 9 mTorr** | |
|  | RMS: 10.61 nm  |
| **(e) 11 mTorr** | |
|  | RMS: 11.45 nm  |

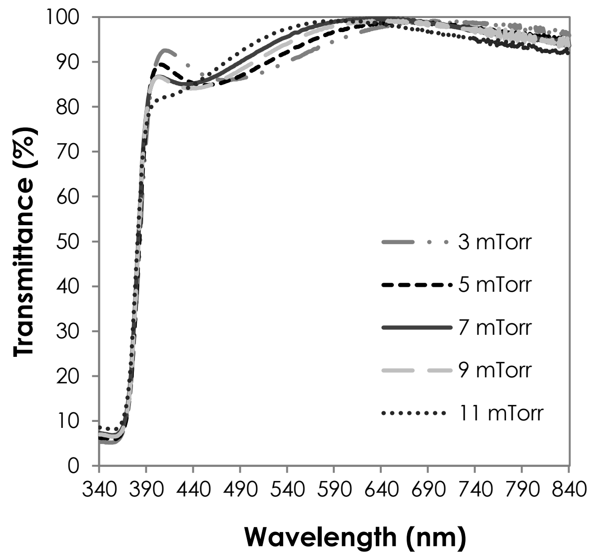



**Figure 4** Optical transmittance spectra of ZnO films deposited on plastic substrate at different sputtering pressure

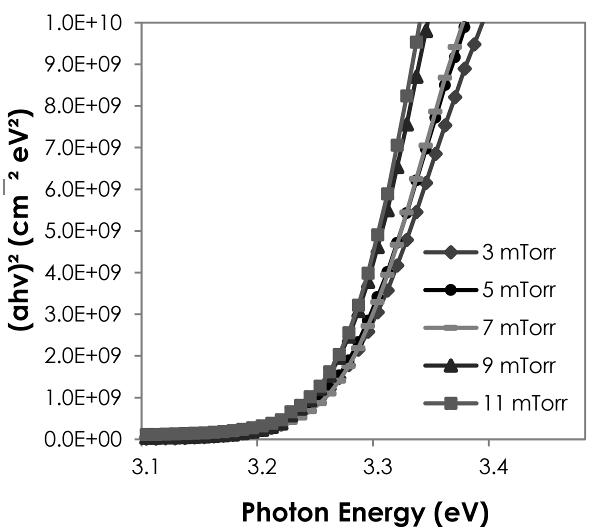



**Figure 5** Plot of graph $(\alpha h\nu)^2$ versus photon energy for ZnO films deposited on plastic substrate at different sputtering pressure

## 4.0 CONCLUSION

ZnO thin films were successfully deposited on the plastic substrate by using RF powered magnetron sputtering method. The film sputtering pressure dependence of structural and optical properties of the plastic substrate held at room temperature was studied. From the X-ray diffraction analysis revealed that the grain size of the films increase and the crystallinity of the films are improve with the increasing of sputtering pressure. From the surface Profilometer studies show that the ZnO thin film thickness decrease, the granular structures enhanced and the surface roughness increase as the sputtering pressure increase. The optical studies revealed that the optical transmittance and band gap increase with the sputtering pressure. High average transmittance of over 80 % for all the thin films is observed and the estimated value of optical band gap is within 3.27 eV-3.29 eV range.

## Acknowledgement

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