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GROWTH CHARACTERISTIC OF GZO FILM FABRICATED BY RF MAGNETRON SPUTTERING

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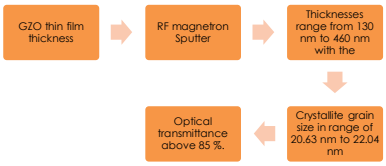
Graphical abstract



Abstract

This paper investigate the dependence of film thickness onto characteristic of Gallium doped Zinc Oxide (GZO). GZO films were deposited on a glass substrate by RF Magnetron Sputtering using GZO ceramic target with 99.99% purity. Thicknesses were altered by varying the deposition time from 10 min to 50 min meanwhile the sputtering power, argon flow and target distance were fixed in order to investigate the influence of film thickness to the growth characteristic, structural, optical properties and surface morphology of the films. Sputtering was performed with RF power of 100 watt and the argon flow was set at 10 sccm. GZO thin films on various thicknesses range from 130 nm to 460 nm were successfully deposited onto glass substrate with the crystallite grain size in range of 20.63 nm to 22.04 nm with the optical transmittance above 85 %.

Keywords: GZO, RF sputtering, growth characteristic, structural properties, optical properties, surface morphology

Abstrak

Kajian ini menyiasat kesan ketebalan filem nipis Galium Zink Oksida (GZO). GZO difabrikasi di atas substrat kaca menggunakan RF Magnetron Sputtering. Ketebalan filem nipis diubah mengunakan tempoh masa fabrikasi filem nipis yang diubah ubah dari 10 minit ke 50 minit, sementara itu kuasa sputtering (100 Watt) , aliran gas Argon (10 sccm), dan juga pemboleh ubah lain dimalarkan bagi menyiasat kesan ketebalan filem nipis ke atas ciri ciri fizikal filem nipis. Filem nipis GZO yang berjaya di fabrikasi dengan ketebalan di antara 130 nm hingga 460 nm mempunyai saiz kristalit di dalam kadar 20.63 nm sehingga 22.04 nm dengan kadar pemindahan optik di atas 85 %.

Kata kunci: GZO, RF sputtering, ciri filem, struktur filem, pemindahan optik, ciri permukaan

1.0 INTRODUCTION

ZnO based thin films have attract a great interest nowadays in semiconductor materials field because of its inexpensive and environmental friendly in term of non toxic properties compared to Indium Tin Oxide (ITO), high transparency in the visible and near ultraviolet spectral region large band gap and high exciton biding energy of 60 meV and it is suitable for semiconductor devices such as solar cells, light emitting diode (LED), and transparent thin film transistor [1-6].

ZnO as a material that have a promising future in semiconductor technology is always attract researcher looking for a way to cover the ZnO imperfection in order to build a perfect ZnO material. Performance of ZnO thin film can be enhanced by introducing a small number of suitable replacement atoms called impurities into semiconductor lattice with a process called doping and by adding dopant

it is believed can alter its electrical and structural properties of ZnO [7].Superiority Gallium when used as dopant for n-type Zinc Oxide semiconductor are high oxidation resistance during deposition, exhibit better electronics stability in humidity also suitable for stabilization of lattice system [8–10]. By adding dopant also will improve the electrical conductivity of Doped Zinc Oxide, where when the Zn^{2+} ions replaced with other ions that have higher valence electrons to act as a shallow donor for example $In^{3+,}$ Al^{3+} and $Ga^{3+.}$ In this research , Ga^{3+} is chosen among others due to the Zn and Ga similar tetrahedral radii that would contribute to the small lattice distortion and minimal strain when the substitutions in doping process occur [10].

Several techniques have been proposed for fabrication of GZO films such as sol gel method ,spray pyrolisis, metal-organic chemical vapor deposition (MOCVD), pulsed laser deposition (PLD) ,dip-coating ,hydrothermal method and also magnetron sputter [5], [11–14].

Among of that techniques magnetron sputtering is chose due to the ability produced high quality thin film with a high density and high adhesion and can be obtained at low substrate temperature with good uniformity of the film thickness in a large area [15].This study will discussed the dependence of film thickness onto characteristic of GZO films.

2.0 MATERIALS AND METHODS

The GZO thin films were deposited onto glass substrate (7.62 cm x 2.54 cm x 0.10 cm) using radio frequency sputtering technique. GZO ceramic target with 99.99% purity from advanced technology material was used as sputter target material. Prior to the deposition, the substrates were cleaned using an Ultrasonic Branson 3200 Cleaner by immersing the substrate in repeatedly three times in distilled water, ethanol, acetone and isopropyl alcohol respectively for 3 minutes and then the substrate was rinsed using distilled water before dried out using dry nitrogen gas. Sputtering was performed with RF power of 100 watt and the argon flow was set at 10 sccm. Thicknesses were altered by varying the deposition time from 10 min to 50 min. The thicknesses, surface morphology, structural and optical properties of GZO film were studied by Profilometer NanoLab 550, XRD (X-ray Diffraction Philips Expert Pro) and UV-Vis spectrometer Lambda 25 respectively. Crystallite grain size and the intensity of thin film were determined using the XRD data obtained. The optical properties of different thicknesses thin films were characterized using UV-Vis Spectrophotometer Lambda EZ210 using wavelength range from 340 nm to 900 nm. The transmittance, absorption coefficient and energy gap values for each thin film were calculated. The Root Mean Square (RMS) for film was calculated to analyzed film surface morphology.

3.0 RESULT AND DISCUSSION

3.1 Growth Characteristics

Thickness and growth rate of GZO films are influenced by the functions of time (Figure 1). As shown in Figure 1, the thicknesses of films are increased in line with the deposition time. GZO thin films on various thicknesses range from 130 nm to 460 nm were successfully deposited onto glass substrate. In the first 10 minutes, the growth rate of films decreased significantly as the deposition time increased. Beyond 20 minutes, the growth rate starts to decrease slowly and almost saturated. The behavior of growth rate of GZO films is related with the structural properties where the grain size of films influences the growth rate of GZO films. In early deposition, the crystallite grain sizes for the films are loose and porous creating an enough area for deposition process occurs but as the deposition time increase, the crystallite grain size for GZO increase. Therefore, the films surface become densely packed hence slows the growth rate of GZO films [9].

3.2 Structural Properties

GZO thin films were successfully deposited on glass substrate (Figure 2) where all the peak observed at the range 33.75 whereas indicate GZO thin film prepared by RF sputtering are polycrystalline with preferential (0 0 2) orientation. Other peak have been observed at (0 0 4) orientation when the deposition time exceed 50 minutes similar to the XRD peak pattern for standard ZnO (JCPDS 36-1451) [16]. No peak of Ga compound observed in XRD pattern suggesting that Ga^{3+} ions filled or substitutes the Zn^{2+} site in ZnO lattice or either Ga^{3+} ions occupy the interstitial sites of ZnO lattice structure [17]. The (0 0 2) peak intensity of films was found showed gradual changes from lower intensity to strong intensity as the film thicknesses increased. The strongest intensity obtained for the GZO deposited at thickness of 365 nm deposited for 40 min. As the thicknesses increased, it is found that the position of 2Θ peak shifted to the higher angle. The changes of peak suggest that there are tensile strains along c-axis direction of the films.

The slightly changes of GZO peak locations also implied that the tensile strains between films are smaller which will contribute to the smaller lattice mismatch [8] suggesting that the addition of Ga^{3+}did not modified the lattice structure of ZnO. The gradual changes of (0 0 2) peak intensity of films were correlated to the crystallite grain size of GZO films where the crystallite grain size has been obtain by using Scherrer's formula (1).

$$D = \frac{0.94\,\lambda}{\beta\,Cos\,\theta} \quad (1)$$

Where X-ray wavelength, λ= 1.54056 Å, β is the FWHM of ZnO diffraction peak, and θ is the Bragg

diffraction angle. The variations of crystallite grain size and fully width half maximum (FWHM) as the functions of time illustrated in Figure 3. The correlation between FWHM and crystallite grain size shows as the films thicknesses increased from 130 nm to 365 nm, value of FWHM decreased, hence the crystallite grain size increase. The result suggesting enhancement of crystalline structure of GZO films obtained at 365 nm thicknesses deposited for 40 min which is in good agreement with the intensity value as shown in Figure 2 where the largest crystallite grain size of GZO films obtained is 22.65 nm. The enhancement of crystallite size influenced by film thickness occur due to the value of lattice constant that decrease as the deposition time increase [18]. Better quality of thin films with a large crystallite grain size will improve the electrical properties of films where the finding in this experiment can be a thickness reference in the future.

3.3 Optical Properties

GZO films fabricated by RF sputtering method shows great optical transmission spectra whereas the average transparency of films was above 80 % as shown in Figure 4. The transparency were differ in different thickness but still show great transparency of film even at highest films thickness of 460 nm. The good optical transmissions properties implied that GZO films are suitable as transparent conductive oxide (TCO) materials. Several interference patterns have been observed suggest that GZO films have a good adhesion contact with glass substrate and also indicated the smooth surface of the GZO films therefore may extend the lifespan of electronics devices with lower possibility to flake off during manufacturing process. The value of optical band gap are obtain from Tauc's equation for direct optical band gap semiconductor as in equation below [19]:

$$(\alpha hv) = A\,(hv - Eg)^{½} \qquad (2)$$

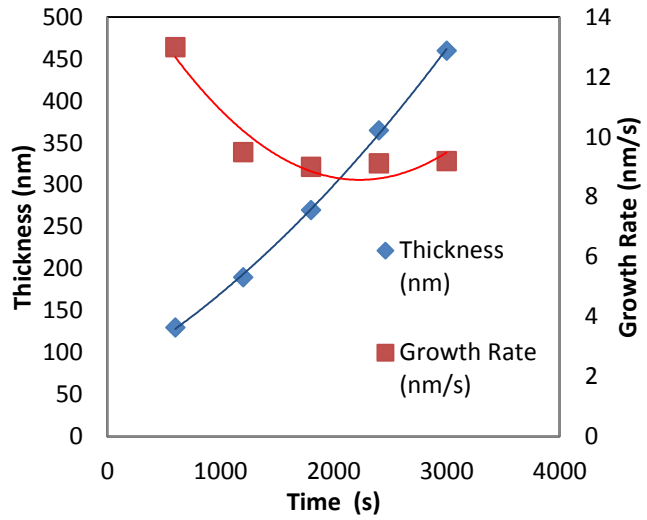


Figure 1 Variation of thickness and growth rate of GZO films by the functions of time

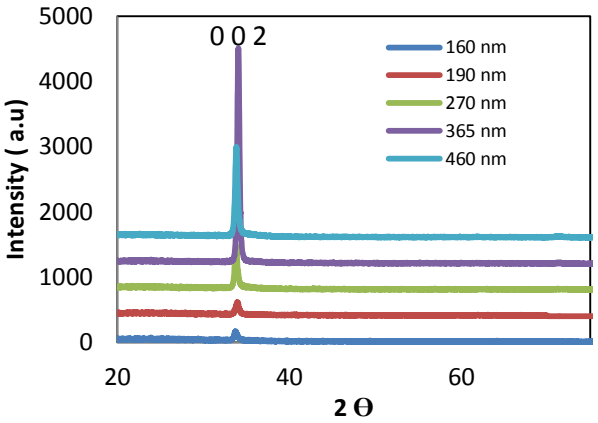


Figure 2 X-ray diffraction (XRD) pattern for GZO thin films at different thickness

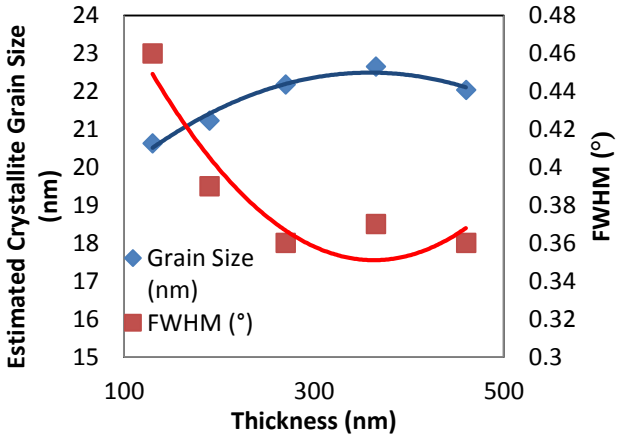


Figure 3 Variation of crystallite grain size and fully width half maximum (FWHM) at different thickness

Where Eg is the energy band gap, α is the absorption coefficient, hv is the incident photon energy and A the edge width parameter. The value of energy band gap Eg evaluated from the extrapolating linear part of the curve obtain from the graph $(ahv)^2$ versus Photon energy (eV). The estimated optical band gap value for the GZO films is within 3.30-3.50 eV range as shown in Figure 5, complement the XRD result that shows only ZnO phase have been successfully deposited into the glass substrate. The value of optical band gap of GZO films were changes as the thicknesses changes. The changes of optical band gap of GZO at different thickness are due to the increase in localized density of states near the band edges and in turn reduce the value of optical band gap of GZO as the thickness increased. The behaviour of optical band gap also correlated with the estimated crystallite grain size of GZO films at different thicknesses were as the particle size increase, the lattice constant also increase hence lower the optical band gap of GZO films at higher thickness which is in agreement with the previous research [20].

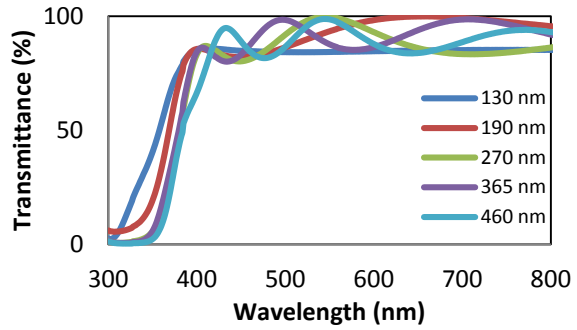


Figure 4 Optical transmittance against wavelength for GZO films at different thickness

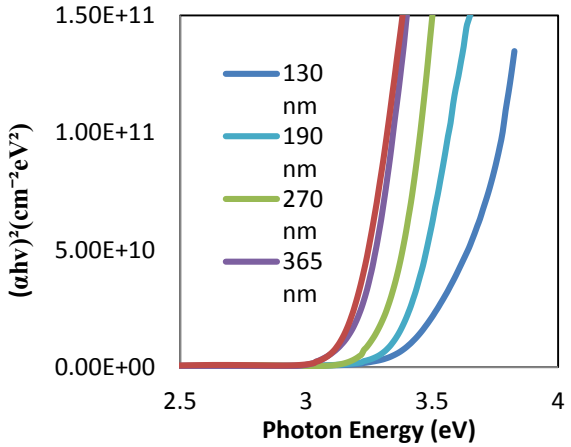


Figure 5 Plot of graph $(\alpha hV)^2$ versus photon energy for GZO films at different thicknesses

3.4 Surface Morphology

The comparisons of surface morphology for GZO obtain from NanoLab 550 Profilometer 3D image films at different films thicknesses are shown in Figure 6. For each data, the surface roughness in term of root-mean-square (RMS) is measured. RMS roughness value of the films can influence the performances of semiconductor devices. The studies revealed a strong influence of thickness on surface morphology of GZO films. The RMS roughness values of the GZO film were found to be in range of 10.2 – 17.9 nm that is increases as the thickness increases. The behavior of the RMS value is influenced by the enlargement of crystallite grain size of thin film which is noticed in Figure 6 where at 130 nm thickness, the GZO film surface were loose and porous and then as the thickness increase, the surface became more densely packed due to the enhancement of crystallite grains size of GZO films hence increase the roughness of GZO films which is similar with the previous research [21]. The increases of RMS roughness value of GZO films agrees well with the increases of estimated grains size from the XRD data shown earlier in Figure 3.

4.0 CONCLUSION

GZO thin films deposited at different thickness were successfully deposited onto glass substrate. Analysis of GZO films structure revealed all GZO films were polycrystalline with the preferential orientation of (0 0 2).The estimated grain size of GZO films were increase as the thickness increase indicated the improvement of crystal quality of films. The optical transmission pattern analysis shows fully transparent films with high transparency achieved with the optical band gap value for the GZO films is within 3.30-3.50 eV range. The studies revealed a strong influence of thickness on surface morphology of GZO films. It is concluded the growth characteristic, structural, optical and surface morphology of GZO films are greatly dependent on films thicknesses.

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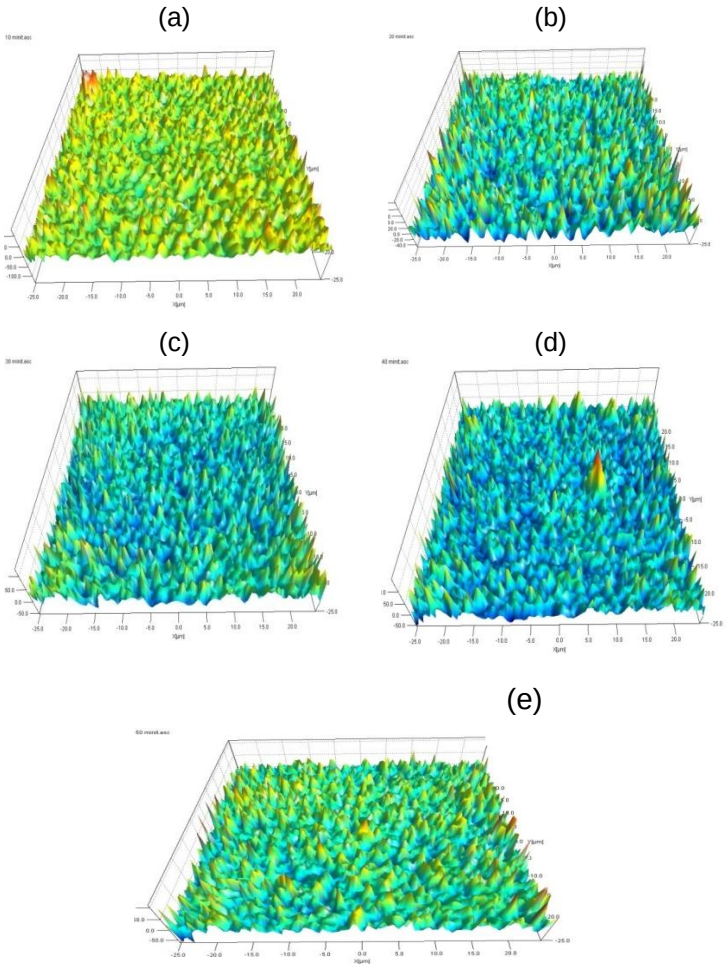

Figure 6 Comparison of surface morphology of GZO (a) 130 nm, (b) 190 nm, (c) 270 nm, (d) 365 nm, and (e) 460 nm films obtained from Profilometer 3D image

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