Oxygen Effect in Annealing Process of Aluminium Doped Zinc Oxide Films

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EDUCATUM JSMT Vol. 5 No. 2 (2018)
ISSN 2289-7070 / eISSN 2462-2451 (33-39)
https://ejournal.upsi.edu.my/journal/EDSC

Oxygen Effect in Annealing Process of Aluminium Doped Zinc Oxide Films

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Abstract

Al doped ZnO (ZnO:Al) thin films was grown on corning glass substrate using dc magnetron sputtering. ZnO:Al thin film grown with plasma power 40 watt and 500 mTorr argon pressure for 120 minutes. Film was grown annealed on oxygen atmosphere with different pressure for 20 minutes. The crystalline of ZnO:Al film deposited has an hexagonal structure. The crystallites preferred orientation shift gradually from (002) to (101) direction as the pressure of oxygen increases. Transmittance in the visible regions increases with increasing pressure of oxygen about 81,27%. The optical characterization indicated that the band gap shift toward lower energies with increasing pressure of oxygen.

Keywords: Aluminium doped zinc oxide, Annealing, Oxygen, Structural properties, optical properties

INTRODUCTION

Zinc oxide (ZnO) is a versatile II-IV compound semiconductor material, it is a functional material with wide and direct band gap of 3.37 eV at room temperature with exciton binding energy (60 MeV) [1]. ZnO is an *n-type* semiconductor of wurtzite crystal structure [2]. Transparent conducting oxide are widely used in microelectronic devices, light emitting diode, thin films, antireflection coating for transparent electrodes in solar cells [3], and gas sensors in surface acoustic wave devices [4], varistors, spintronic devices, and lasers [5].

In addition, ZnO films doped with Al, Ga, or In have low electrical resistivity and high optical transmittance for optoelectronic and electronic devices, such as flat panel display, solar cells, and led emitting diodes [6]. The Al-doped ZnO (AZO) transparent conducting film shows the lowest resistivity among impurity doped ZnO films [7]. Recently, ZnO has considered as a potential substitution material for ITO and SnO_2 widely used as transparent conducting materials, owing a number of encouraging advantages namely low cost and stability under reduced hydrogen atmosphere [8,9].

ZnO:Al films can be prepared by several techniques including metal oxide chemical vapour deposition (MOCVD) [10], sol-gel-dip-coating [11], buffer assistend pulsed laser deposition [12], pulsed laser deposition [13], electrodeposition [14], chemical spray pyrolysis [15,16,17], and sputtering [18]. In this work was used DC Magnetron sputtering method to deposit Al: ZnO thin films.

The study investigated the effect of annealing process in oxygen ambient with different pressure on structural and optical properties of Al-doped ZnO films as a function of oxygen ambient. A ZnO:Al (Al_2O_3: 3 wt%) target was used and AZO films were grown at 400°C. the films were annealed at 300°C with oxygen pressure ranging from 0 mTorr to 200 mTorr in steps of 50 mTorr.

EXPERIMENTAL DETAILS

Al doped ZnO films were deposited on the glass substrate by DC magnetron sputtering using an AZO ceramic target (AL_2O_3: ZnO = 3: 97 wt%). The glass substrate were ultrasonically cleaned in an ethanol/acetone solution and then rinsed with nitrogen blow gas. The deposition AZO layers was performed in an argon (purity: 99.99%) atmosphere. The Ar flow was maintained at 500 mTorr and the DC power was controlled at 40 W. substrate temperature kept at 400°C for 120 minutes. Then, the AZO films are annealed in oxygen ambient. The annealed temperature at 300°C and pressure of oxygen of the films are at 0, 50, 100, 150, and 200 mTorr for 20 min. Optical transmittance was measured using a UV-VIS-IR spectrophotometer in the range of 300-800 nm. Conventional $\theta - 2\theta$XRD studies on films were carried out in Rigaku diffractometer using Cu Kα radiation to investigate the crystallinity and crystal orientation of the films.

RESULT AND DISCUSSION

Fig. 1 shows the XRD diffractogram of ZnO:Al thin films as deposited and annealing in oxygen ambient at 0 mTorr, 50 mTorr, 100 mTorr, 150 mTorr, and 200 mTorr pressures. Two peak appear on the XRD pattern for all films deposited, they correspond to the (002) and (101) directions assigned to wurtzitehexagonal structure. The hexagonal structure indicated that films growth is achieved along the *c*-axis perpendicular to the substrate surface [19], with increasing pressure oxygen, the intensity of diffraction peak decreases. This is due to the kinetic energy reduction of reactive species in the chamber when the pressure of oxygen is increased, which limits the atom surface diffusion [20].

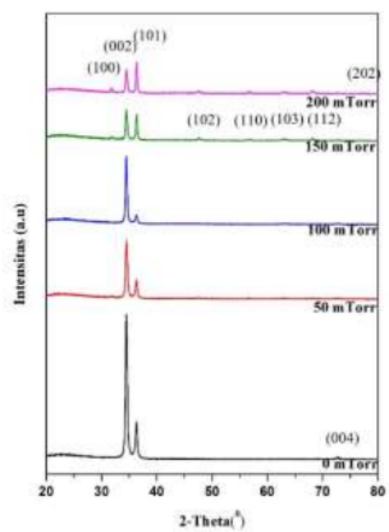

Figure 1. X-Rays diffraction patterns of ZnO:Al films with different oxygen pressure on annealing process

EDUCATUM JSMT Vol. 5 No. 2 (2018)
ISSN 2289-7070 / eISSN 2462-2451 (33-39)
https://ejournal.upsi.edu.my/journal/EDSC

The preferred peak diffraction showed Fig. 1 is peak on (101) direction. It is extremely interesting to study when give variation treatment on oxygen pressure of annealing process. Intensity of (101) peak was decreased when oxygen pressure was increased of 100 mTorr at annealing process. When the oxygen pressure on the annealing process was increased from 150 mTorr to 200 mTorr, peak intensity of (101) direction also increases.

On oxygen pressure 200 mTorr the intensity on (101) peak is higher than intensity on (002) peak. It is the consequence of increasing on substrate or film temperature of annealing process [21]. The growth direction of film on (002) peak has disappear and shift by other direction on diffraction spectrum (101) peak [22]. ZnO:AL thin film on (101) peak orientation can be applied as photo anode single layer and double layer pada Dye-Sensitized Solar Cell (DSSC) [23].

Diffraction peak on (101) orientation plane shown peak with hexagonal phase, uniform of grain growth, and perpendicular to substrate surface. The growth of (101) orientation plane indicated that ZnO:Al thin film is structured along a-axis [24].

The Result of XRD characterization is used to determined full width at half maximum (FWHM) and crystal size. The fine crystal is crystal that has small FWHM with high crystal size and decrease on grain boundary [25]. FWHM of (101) peak is smaller when oxygen pressure on annealing process increase from 100 mTorr to 200 mTorr. The smallest FWHM on ZnO:Al thin film with 100 mTorr oxygen pressure, it is 0,28°. The increasing of oxygen pressure effect on oxygen atoms had absorbed is much more and fill oxygen vacancy on ZnO:Al film. The largest crystal size on ZnO:Al thin film with 200 mTorr oxygen pressure, consistently with the smallest FWHM based on research of Hsu et al., (2011). The correlation between FWHM value and crystal size with oxygen pressure on annealing process can be showed on Fig. 2.

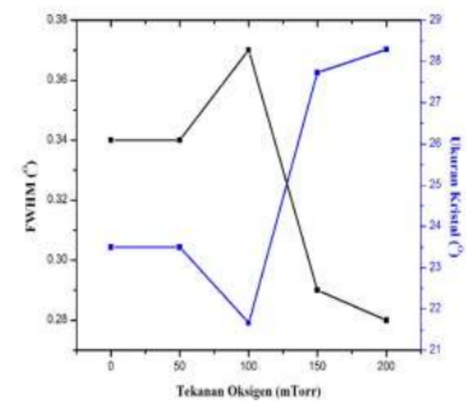

Figure 2. FWHM and crystal size of (101) peak ZnO:Al films with different oxygen pressure on annealing process

The shift crystal size and FWHM value of film is indicate lattice strain level on thin film. Lattice strain in film showed of stress on film. Stress on film influence to crystal quality that has produced. Higher stress that occur on film made better quality of thin film that has produced.

The Opticcharacteristic of thin film characterized by UV-Vis Spectrophotometer. The result of characterization is optic transmittance graph of ZnO:Al thin film that has growth, showed on Fig. 3.

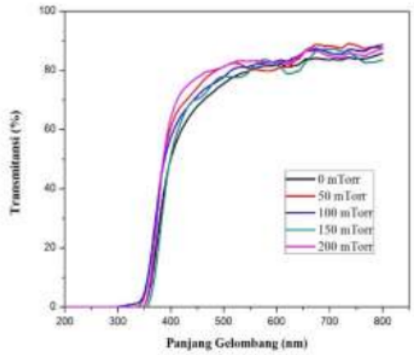

Figure 3. Optical transmittance of ZNO:Al film annealing in oxygen with different pressure

Analysis of UV-Vis showed that transparent characteristic related to film quality that has formed and can be influenced by crystal structure and grain size of thin films. The average of ZnO:Al thin film transmittances with oxygen pressure variation on annealing process with wavelength ranges 400 nm – 800 nm as 81,27% on visible light spectrum ranges. The Graph correlation between transmittances and variation of oxygen pressure while annealing process showed Fig. 4.

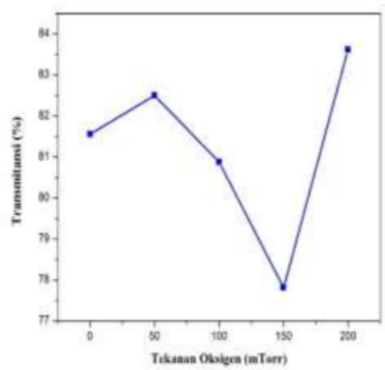

Figure 4. Graph correlation between transmittances and variation of oxygen pressure while annealing process

Transmittances increment showed there are increasing crystallinity and decreasing roughness of surfaces [26]. It is summarized that increasing oxygen on ZnO:Al thin film annealing process can increase on thin film transmittances that has been growth.

Analysis of UV-Vis can be used to determined band gap energy of ZnO:Al thin film with variation of oxygen pressure on annealing process. Band gap energy is gotten from linier extrapolation toward graph of correlation absorption coefficient squared $(\alpha h\upsilon)^2$ with foton energy $h\upsilon$ (eV) to $(\alpha h\upsilon)^2 = 0$ as showed on Fig. 5.

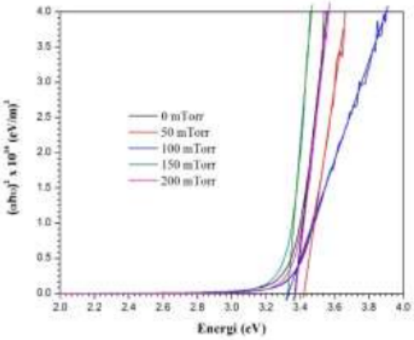

Fig 5. Variation of $(\alpha h\upsilon)^2$as a function of foton energy hυ

Based on extrapolation was gotten band gap energy toward to variation of oxygen pressure on annealing process as showed on Table 1.

Table 1. Band gap energy toward to variation of oxygen pressure on annealing process

Oxygen Pressure (mTorr)	*band gap Energy* (eV)
0	3,36
50	3,40
100	3,32
150	3,29
200	3,36

Based on Table 1. Seen that band gap energy different however the differences is not significant. Band gap energy of ZnO:Al with 0 mTorr oxygen pressure is 3,36 eV, 50 mTorr oxygen pressure is increase to 3,40 eV. The increment of band gap energy coincide with increment of carrier concentration on thin film that has produced [19]. On 100 mTorr and 150 mTorr of oxygen pressure, band gap energy is decrease to 3,32 eV and 3,29 eV, whereas on 200 mTorr pressure the band gap energy is increase to 3,36 eV.

Band gap energy of thin film that has been produced is still on ranges of TCO material, so the thin film that has produced can be applied as TCO material and window layer on solar cell. It is based on TCO material that has ranges of band gap energy between 2,5-4,5 eV [27].

CONCLUSION

ZnO:Al thin film was deposited in corning glass substrates with DC Magnetron Sputtering method successfully. The result of ZnO:Al thin film XRD characterization has showed that thin film which has deposited has hexagonal structure with dominant diffraction peak match on (002) and (101) orientation plane. (002) orientation plane is used as windows layer application on solar cell and (101) orientation plane can be applied as photo anode single layer and double layer on Dye-Sensitized Solar Cell (DSSC).

The result of ZnO:Al optic characterization used UV-Vis shown that transmittances of ZnO:Al thin film with variation of oxygen pressure on annealing process is 81,27%. Band gap energy of ZnO:Al with variation of oxygen pressure that has been produced on annealing process is different, band gap energy differences is not significant and included on ranges of TCO band gap energy, then film that has produced can be applied as TCO material or window layer on solar cells.

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