FABRICATION AND CHARACTERIZATION OF Cu_2O/ZnO THIN FILMS FOR pn HETEROJUNCTION DEVICES

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ABSTRACT. *This paper described the fabrication of Zinc Oxide (ZnO)-Cuprous Oxide (Cu_2O) heterojunction thin films using RF-powered Magnetron Sputtering System. The deposition parameters were controlled to produce the films with the thicknesses in the range of 100 nm to 500 nm. During deposition, the RF power and the argon flow are fixed at 100 Watt and 10 sccm, respectively. Structural and optical properties were studied by X-Ray Diffraction Method and UV-VIS Spectrometer. The electrical properties were studied by IV SourceMeter. The grain sizes of both thin films increasing while the thickness increase. The band gap of ZnO thin films range from 3.25 eV -3.27 eV and for Cu_2O thin films range from 2.00 eV – 2.15 e V. All Cu_2O/ZnO thin films show ideal diode properties.*

KEYWORDS: ZnO, Cu_2O, thin film, heterojunction device

INTRODUCTION

ZnO based thin films have attracted a great interest nowadays in semiconductor materials field because of its inexpensive and environmental friendly as compared to Indium Tin Oxide (ITO), high transparency in the visible and near ultraviolet spectral region, large band gap and high exciton binding energy of 60 *meV* and also its suitability for Transparent Conductive Oxide (TCO's) devices (Park, 2006). Numerous techniques have been proposed for fabrication of Cu_2O/ZnO 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 (Senthilkumaar *et al.*, 2008). Among these techniques, magnetron sputtering is chosen due to its ability to produce 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 (Nam *et al.*, 2014).

In this study the main materials that used in fabrication of heterojunction devices thin films are Zinc Oxide (ZnO) and Cuprous Oxide (Cu_2O). Both of these materials receive positive review from lots of researchers. Cu_2O/ZnO is an attractive all-oxide candidate for low-cost photovoltaic applications. The materials are abundant, non-toxic and relatively stable. ZnO is a natural n-type semiconductor with an energy gap of around 3.37 eV (Vinodkumar *et al.*, 2010). ZnO has been used to fabricate many crucial devices such as heterojunction devices window (Craciun *et al.*, 1995), light emitting devices (Yamamoto *et al.*, 1980) and photo detectors (Singh *et al.*, 2007).

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ZnO has drawn the attention of researchers on its unique properties such as high electrochemical stability, resistivity control, and good transparency in the visible range with a wide band gap and the absence of toxicity and abundance in nature (Singh *et al.,* 2007).

Cu_2O is a semiconductor with a band gap of 2.0 eV (Tsur and Riess, 1999). Although not optimal (1.5 eV), its band gap nearly ideal when used as a top cell or absorber layer in hetero junction heterojunction devices. It is known as suitable material for solar cell due to its direct band gap and optical absorption coefficients. Cu_2O also has been used as cathode material for micro batteries (Souza *et al.,* 2006), random access memories (Dong *et al.,* 2007), high temperature superconductors (Li *et al.,* 1991) and gas sensor (Samarasekara *et al.,* 2006). This study discussed the dependence of thickness on the structural, optical and the efficiency of Cu_2O/ZnO heterojunction devices prepared by RF powered magnetron sputtering system on a glass substrate.

METHODOLOGY

Cu_2O/ZnO heterojunction devices films were deposited onto glass substrate (7.62 cm x 2.54 cm x 0.10 cm) under different argon pressure in a RF powered magnetron sputtering system. Cu_2O and ZnO ceramic target with 99.99% purity were used as sputtering target material. Prior the deposition, the substrates were cleaned using Ultrasonic Branson 3200 Cleaner. The substrate was immersed repeatedly for three times in distilled water, ethanol, acetone and isopropyl alcohol respectively for 3 minutes and then rinsed using distilled water before dried out using dry nitrogen gas. Initially the vacuum chamber of RF magnetron sputtering was evacuated to the base pressure of 1.0 x 10^{-5} to and then the sputtering was performed with RF power of 100 watt at room temperature. The structure of Cu_2O and ZnO thin films were studied using X-Ray Diffraction (Philips Analytical X-Ray). The XRD pattern was obtained and the grain sizes of thin films were calculated using *The Scherrer Formula*.

$$B(2\theta) = \frac{K\lambda}{L\cos\theta} \tag{1}$$

where $B(2\theta)$ is grain size, K is shape factor, λ is X-Ray wavelength, L is line broadening at half the maximum intensity (FWHM) and θ is the incident angle of X-ray. The transmission spectra of Cu_2O and ZnO thin films were obtained from UV-Vis spectrometer Lambda 25. The optical band gap of Cu_2O and ZnO thin films were calculated using *Tauc's Equation deriviation.*

$$\alpha hv = A\left(hv - E_g^{\ 1/2}\right) \tag{2}$$

where E_g is the band gap energy, α is the absorption coefficient, hv is the incident photon energy and A is the edge width parameter. The value of band gap energy, E_g was evaluated from the extrapolation of the linear slope (αhv^2) versus photon energy eV. Lastly the electrical properties of Cu_2O/ZnO film were determined using IV measurement (Keithley 2400 SourceMeter MSU).

RESULT AND DISCUSSION

A. Structural properties

Figure 1 (a) and (b) show the X-ray diffraction pattern of ZnO and Cu_2O thin films respectively at different thickness ranging from 100 nm to 500 nm under room temperature. The findings show that all the ZnO thin films are polycrystalline structure and the peak at about 34° corresponds to the diffraction from the (0 0 2) plane of ZnO. It is indicated that the fabricated thin films have a hexagonal wurtzite structure and preferentially oriented along the c-axis perpendicular to the plane of the substrate. From the XRD pattern, the peak intensity of (0 0 2) is found increased with the increasing of thin film thickness. The highest intensity is observed at thickness of 500 nm.

Figure 1 (b) also shows that the peak intensity is directly proportional to the thin film thickness. At a higher thickness, more than one peak of Cu_2O is detected impled that Cu_2O films fabricated by RF magnetron sputtering is polycrystalline with an orientation of (1 1 1) as the major preferential orientation. The other minor peak obtained is (1 1 0) and (1 1 3).

Figure 2 (a) and (b) show the variations of full width half maximum (FWHM) and crystallite grain size as a function of thickness of ZnO and Cu_2O thin films respectively. It is found that the value of FWHM is inversely proportional with the crystallite grain size. The crystallite grain size of ZnO film are enhanced with the increasing of film thickness.

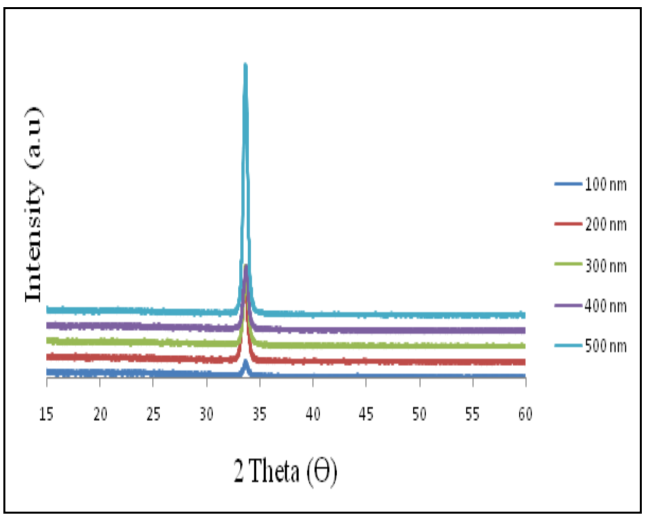


Figure 1(a): X-ray diffraction pattern of ZnO thin films at different thickness(nm).

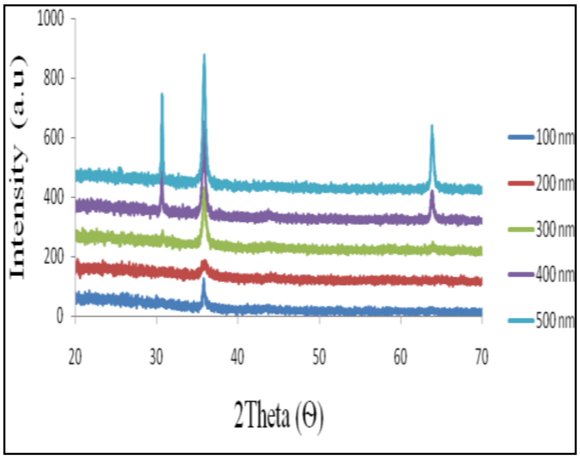
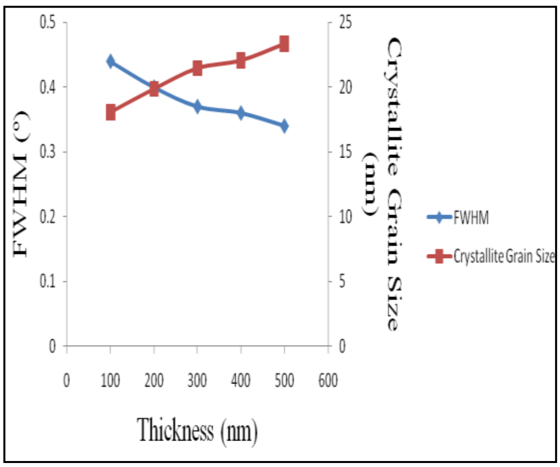
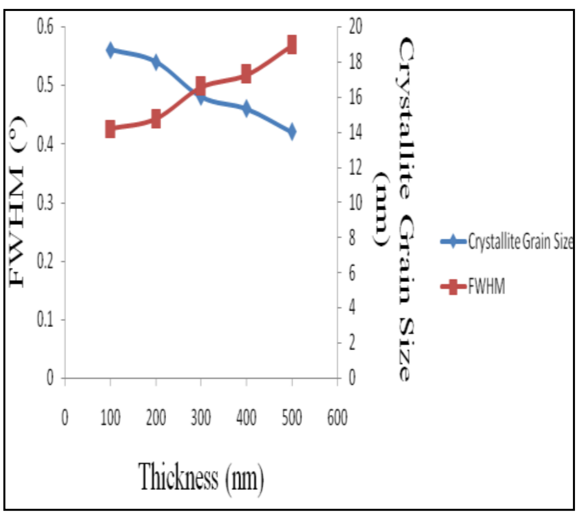


Figure 1(b): X-ray diffraction pattern of Cu_2O thin films at different thickness(nm).



(a)



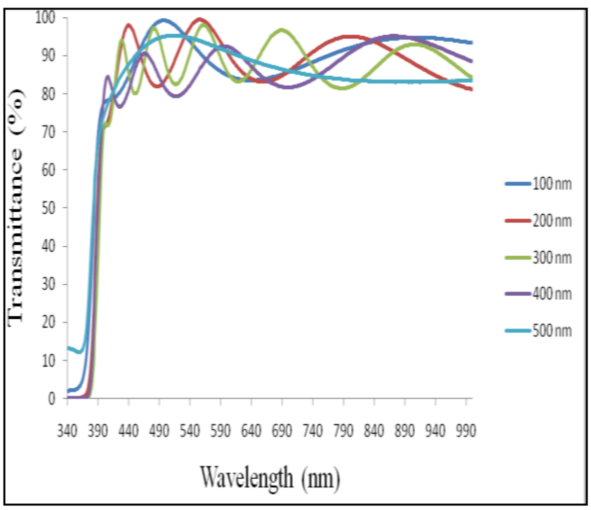
(b)

Figure 2 : Variation of Crystallite Grain Size and Full Width Half Maximum (FWHM) **(a)** Cu_2O and **(b)** ZnO as the Functions of Thicknesses.

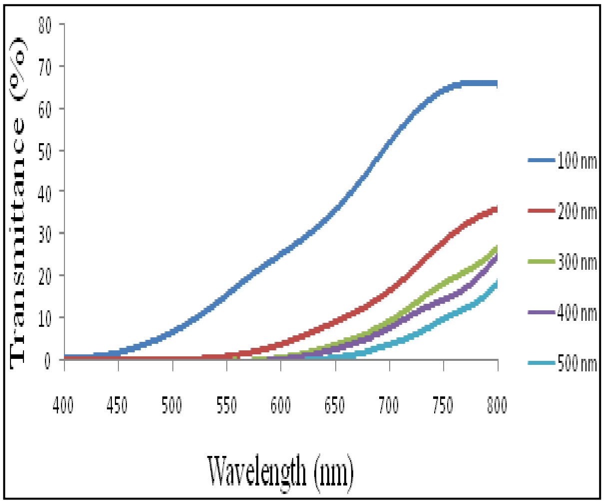
B. Optical Properties

Figure 3 (a) and (b) show the transmission spectrum of ZnO and Cu_2O thin films at different thicknesses. All the ZnO samples show an excellent transparency within the visible light region (Nam *et al.*, 2014) with the transmission rate above 80% paralleled with the previous research (Noda *et al.*, 2013). From the observation, the transmission value of the films do not show significant difference. From the spectra, there was no significant effect of thickness on the transmission. For Cu_2O samples, the transmission percentage is less than ZnO sample. Cu_2O is physically dark and therefore the fabricated thin film is less transparent compared to ZnO thin film.

These result agreed well with previous research works (Ghotbi, 2012). Figure 4 (a) and (b) show αhv^2 against photon energy at different thickness for ZnO and Cu_2O. E_g was estimated from extrapolation graph line and the optical band gap was obtained. The estimated optical band gap for ZnO is in the range of 3.25 eV -3.27 eV. The estimated energy band gap of different thickness ZnO thin films are in good agreement with other works (Willander *et al.*, 2009). The samples of ZnO thin films are having excellent transparency with the optical transmission for all samples above 80%. As for Cu_2O thin films, the energy band gap is in the range of 2.00 eV – 2.15 e V. The values of band gap decrease as the thin film thickness increase.

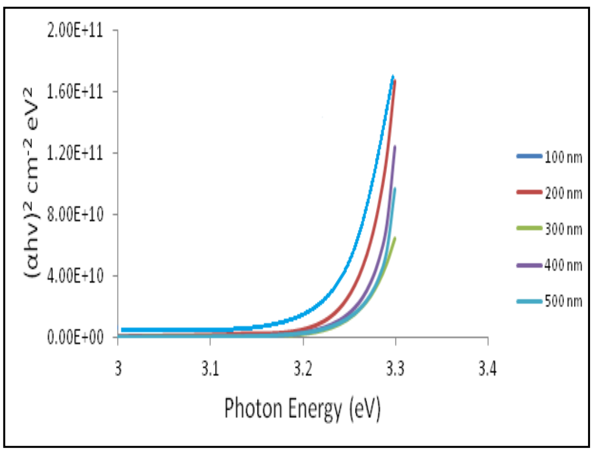


(a)

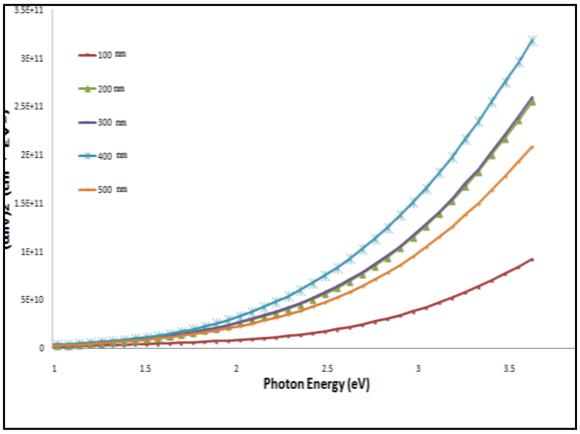


(b)

Figure 3 and: Transmission spectrums of **(a)** ZnO and **(b)** Cu_2O as the Functions of thicknesses



(a)



(b)

Figure 4 : Band gap graph of **(a)** ZnO and **(b)** Cu_2O as the Functions of Thicknesses

C. Electrical Properties

Figure 5 shows the current-voltage (I-V) characteristics of Cu_2O/ZnO p-n heterojunctions at different thickness. All samples show ideal diode properties. As seen in Figure 5, the combination thickness 300nm/500nm of Cu_2O/ZnO thin film establish ideal diode properties which indicating the heterojunction devices efficiency. This situation might occur due to the lowest lattice mismatch between the structure of ZnO/ Cu_2O. Both thin films have sharp peak of XRD pattern, indicate good structural properties of the thin films. The reductions in grains boundary scattering is due to the increment of carriers which improves the conductivity and solar efficiency. (Oztas, 2008).

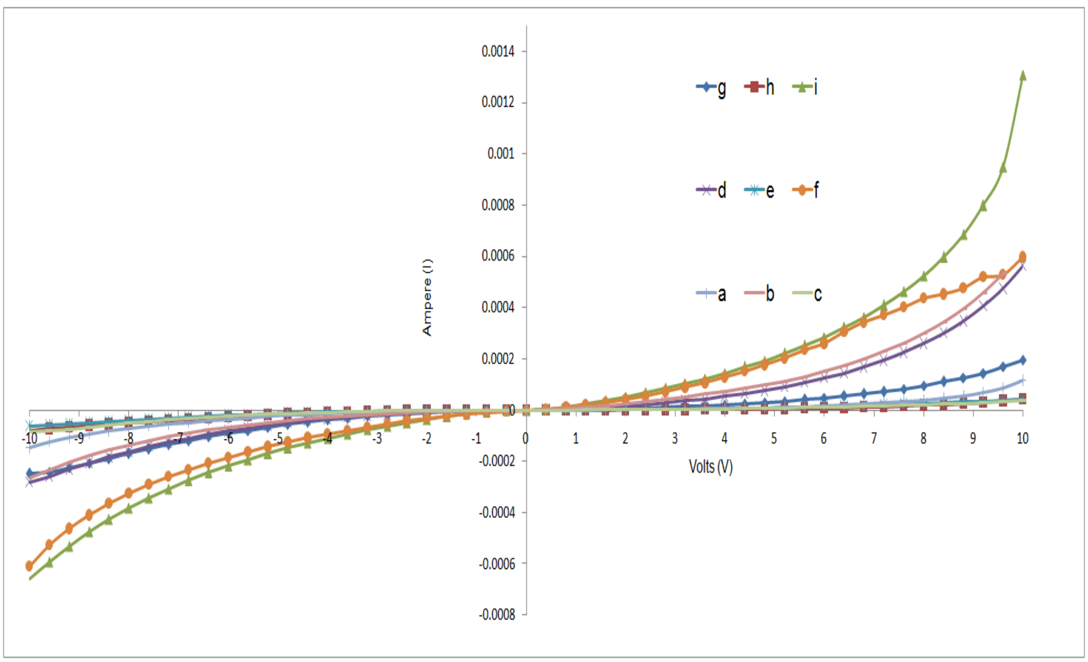


Figure 5: I/V curves of Cu_2O/ZnO p-n heterojunctions

CONCLUSION

Cu_2O/ZnO at different thickness were successfully deposited onto the glass substrate using RF magnetron sputtering technique. The findings reveal that Cu_2O/ZnO thin film is polycrystalline and fully transparent. The average optical band gap energy for the fabricated ZnO film is 3.30 eV and 2.05 eV for Cu_2O film. The highest ideality factor is observed at the Cu_2O/ZnO thin film with both of their thickness at 300 nm.

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