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ORIGINAL ARTICLE

Annealing effects on the structural, electrical and optical properties of ZnO thin films prepared by thermal evaporation technique

A. Zaier [a,b,*], A. Meftah [b], A.Y. Jaber [c], A.A. Abdelaziz [c], M.S. Aida [d]

[a] Department of Pharmacy, Faculty of Medicine, University of Constantine 3, Algeria
[b] LRPCSI, Faculty of Sciences, University of Skikda, BP 26, 21000 Skikda, Algeria
[c] Laboratory of Thin Films and Solar Cells, Department of Physics, Faculty of Sciences, Taibah University, Al Madinah, Saudi Arabia
[d] Thin Films and Interfaces Laboratory, Faculty of Sciences, University of Constantine 1, Algeria

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Abstract Zinc oxide (ZnO) thin films have been prepared on glass substrates at room temperature by thermal evaporation technique using ZnO powders and then are annealed at different temperatures ranging from 200 °C to 500 °C for 2 h in air. The effect of the annealing temperature (T_r) on the structural, optical and electrical properties of the ZnO thin films was studied.

Experimental results show that annealing temperature has an important role in the changes observed in the structural, optical and electrical properties of the ZnO thin films. The XRD measurements confirm that the thin films grown by this technique have good crystalline hexagonal wurtzite structures. The optical transmittance spectra show transmittance higher about ~90% within the visible wavelength region. Hence, the values of the gap are found to be between 3.13 and 3.25 eV. The resistivity values of the films have changed between 2.10^{-3} and 4.10^{-2} Ω cm with annealing temperature.

1. Introduction

Zinc oxide (ZnO) is a wide-band gap n-type semiconductor of the II–VI semiconductor group with a zinc excess in interstitial position. It is currently the subject of numerous studies because of its exceptional properties, such as more elevated chemical and thermal stability even in atmosphere of hydrogen plasma compared to other oxides (SnO_2, ITO), large exciton binding energy of 60 meV at room temperature, non-toxicity and its low price (Volkan et al., 2014; Chrissanthopoulos

* Corresponding author at: Department of Pharmacy, Faculty of Medicine, University of Constantine 3, Algeria.
E-mail address: zaier_abdelhakim@yahoo.fr (A. Zaier).
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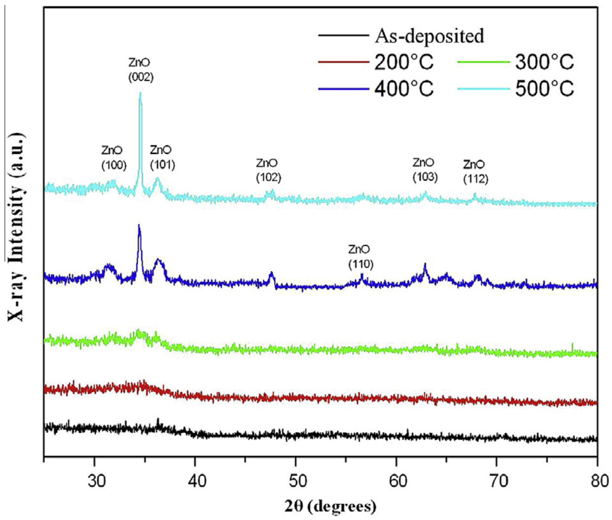

Figure 1 XRD patterns of ZnO thin films annealed at various temperatures.

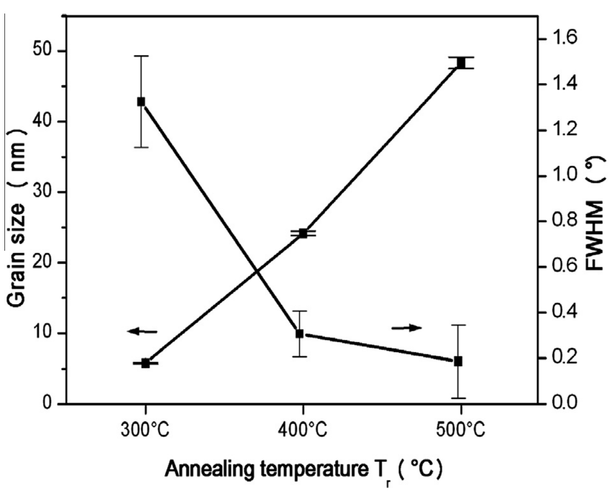

Figure 2 FWHM of the main XRD peak and corresponding grain sizes of ZnO films as a function of the annealing temperature.

et al., 2011; Mimouni et al., 2015), which allow it to be like a material model especially in its extremely various applications (electronics, optics, optoelectronics, conversion photovoltaic) (Chuen et al., 2012; Djurisic et al., 2010; Al-Hardan et al., 2014; Keun et al., 2013). ZnO thin films have been prepared by various techniques such as thermal evaporation (Chrissanthopoulos et al., 2011; Ghislain et al., 2013; Yuvaraj et al., 2008; Feng et al., 2010; Nguyen et al., 2014), Pulsed laser deposition (Raied et al., 2014), molecular beam epitaxy (Zhang et al., 2011), magnetron sputtering (Ismail and Abdullah, 2013; Mosbah and Aida, 2012), sol–gel (Hashim et al., 2013; Linhua et al., 2012), chemical vapour deposition (Hsiao et al., 2013; The-Long et al., 2010; Jeon et al., 2014) and spray pyrolysis (Zaier et al., 2009; Lucio et al., 2006). Thermal evaporation is an interesting technique because of its simple process of deposition, films uniformity and mainly species evaporation controllability. The present

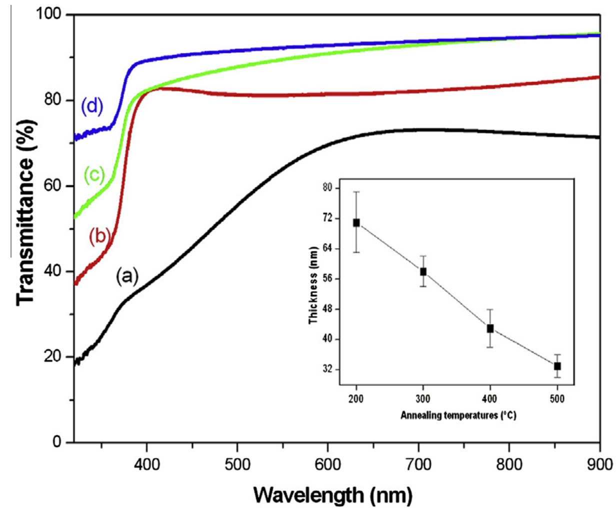

Figure 3 The transmission (%) spectra of ZnO thin film annealed at different temperatures (T_r): (a) 200 °C, (b) 300 °C, (c) 400 °C, and (d) 500 °C. The inset shows the films thickness variation of different annealed temperatures.

work deals with ZnO thin film deposition using thermal evaporation. The structural, optical and electrical properties of the films were studied as a function of annealing temperature.

2. Experimental

ZnO thin films, used in this study, were deposited onto glass substrates at room temperature by thermal evaporation technique using ZnO powders (99.99% purity). The distance between Substrate to source was 6 cm. The base pressure in the deposition chamber was evacuated to 10–5 mbar. The source current was increased slowly (up to 3 A) and the vapour species condenses onto glass substrates. After deposition, these films were followed by thermal oxidation in air using furnace Linn High Therm GmbH (Model LM 312) at various temperatures ranging from 200 °C to 500 °C for a fixed time of 2 h. The films structural characterisation was carried out using X-ray diffraction (ray $K_{\alpha 1}$ of the Cu, λ = 1.5405 Å) system. Films thickness was measured by Dektak profilometry. For the optical properties measurements, we used a double beam Shimadzu 1700 UV–vis spectrophotometer with an integrating sphere in the wavelength range from 200 to 1100 nm. The electrical properties measurements were performed with four probe method.

3. Results and discussion

3.1. Structural properties

Fig. 1 shows the recorded XRD diffraction pattern of ZnO samples deposited by thermal evaporation technique and annealed at different temperatures. This result clearly shows that ZnO thin film is polycrystalline hexagonal wurtzite structure (according to JCPDS card No. 00-036-1451), these results were found by Raied et al. (2014). Strong (002) preferential orientation indicates polycrystalline nature of the thin films ZnO.

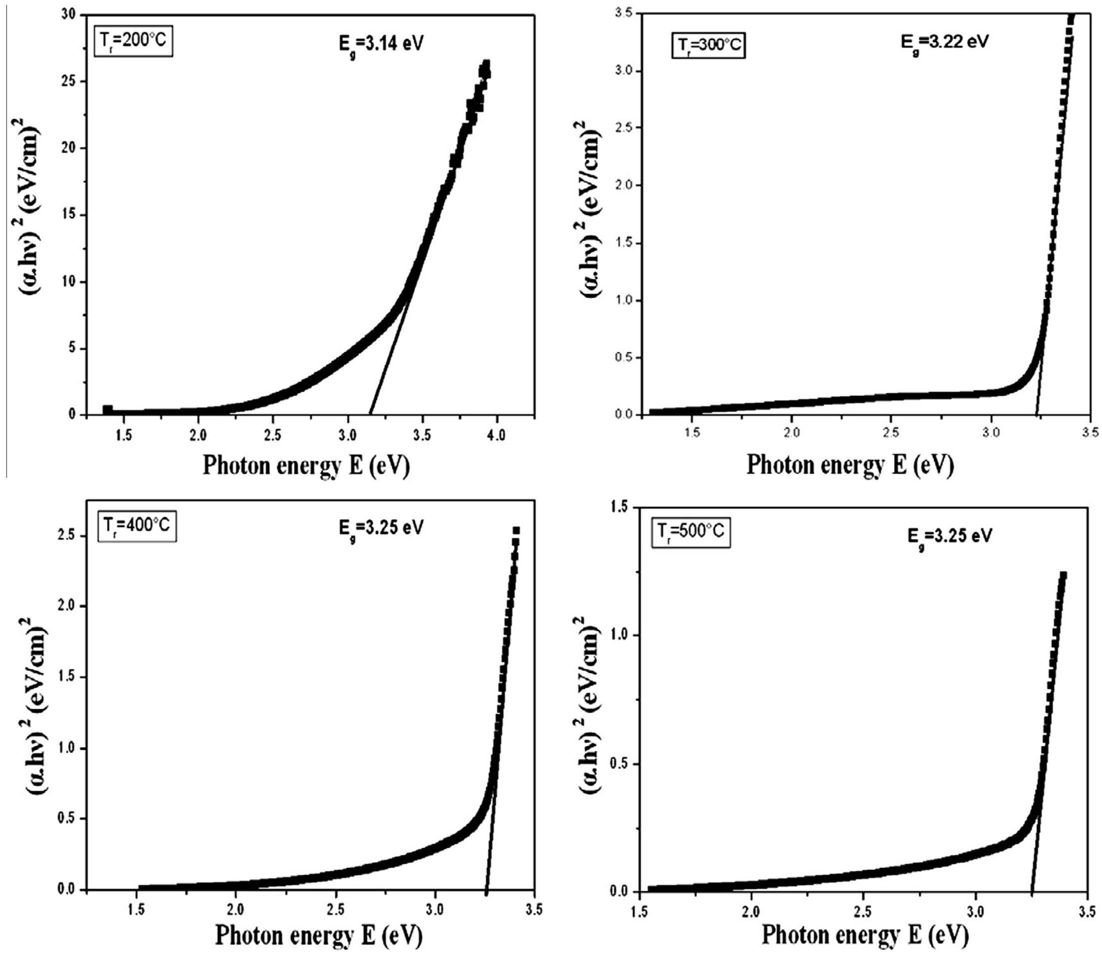

Figure 4 The plot of $(\alpha h\nu)^2$ versus photon energy ($h\nu$) for ZnO thin films prepared at various temperatures (T_r).

The crystallite size of the ZnO thin films was calculated by using the Debye–Scherrer formula (Hosseinmardi et al., 2012):

$$D = \frac{0.9\lambda}{\beta\cos\theta} \tag{1}$$

where λ is wavelength of the incident X-ray (λ = 1.5405 Å), β is the full width at half maximum (corrected for instrumental broadening), and θ is the Bragg's angle.

Fig. 2 shows ZnO diffraction peak (002) full width at half-maximum (FWHM) and mean grain size as a function of the annealing temperatures (T_r). With elevating annealing temperature the film's mean grain size increases, and FWHM of the (002) peak becomes narrower indicating the improvement of the crystalline of ZnO.

3.2. Optical properties

The optical transmittance spectra with wavelengths from 320 to 900 nm ZnO films annealed for different temperatures are presented in Fig. 3. Optical transmission increases according to the annealing temperature from 200 °C to 500 °C until a value of 90% for the films annealing at 500 °C, because of structural homogeneity, crystallinity and the thicknesses of films, as reported by Hamid et al. (2007).

The band gap of the films corresponding to annealing temperature for 200–500 °C was calculated by plotting $(\alpha h\nu)^2$ vs $h\nu$ using the relation (Ayouchi et al., 2003):

$$\alpha h\nu = A(h\nu - E_g)^{1/2} \tag{2}$$

where, α: is the absorption coefficient, $h\nu$: is the photon energy, E_g: is the band-gap. A: is an energy-independent constant.

The energy gap (E_g) value is calculated by extrapolation of the straight line of the plot of $(\alpha h\nu)^2$ versus photon energy ($h\nu$) for the different annealing temperatures, are shown in Fig. 4. The energy band gap increases with the annealing temperature. It varies from 3.13 eV to 3.25 eV for 200 °C to 500 °C annealing temperatures, respectively. These values of the band gap are in agreement with those found by Malek et al. (2014). This change in energy band gap can be attributed to the reduction of the structural defects in films network and also to the increase of grain size with annealing.

3.3. Electrical properties

The resistivity of the films is calculated using the following equation:

$$\rho = \frac{kV}{I}d$$

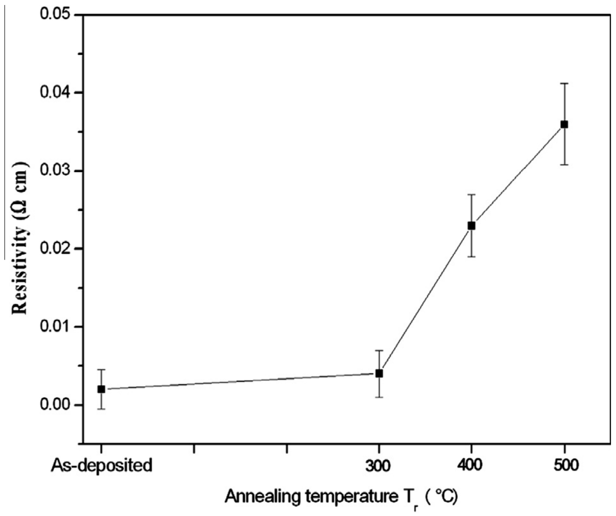

Figure 5 Variation of the electrical resistivity with the annealing temperature.

where: d: is the thickness of the sample, V: is the voltage and I: is the current. K: is a constant dependent on the geometry is equal with $\frac{\pi}{\ln 2}$.

The resistivity variation with annealing temperature of the ZnO films is shown in Fig. 5. As can be seen, the resistivity increases with annealing temperature from 2.10^{-3} to 4.10^{-2} Ω cm for 0 °C (As deposited) and 500 °C respectively. This can be attributed to the increase of oxygen when increasing the annealing temperature that reduces the number of oxygen vacancies (free carrier) in ZnO films. Asghar et al. (2008) and Bouhssira et al. (2006) have reported similar results.

4. Conclusion

In summary, ZnO thin films have been deposited by thermal evaporation method on glass substrates. The films have been annealed in a furnace at different temperatures. The XRD results reveal that the deposited thin film has a good polycrystalline hexagonal wurtzite structure. The size of crystalline (grains) for annealing at 500 °C is about 1.5 nm. The average transmittance in visible region is about ~90% increasing with annealing temperature. The optical band gap of ZnO films is in the range of 3.13–3.25 eV. The more the electrical resistivity of the films increases, the more the annealing temperature rises. Finally, we can say that we have prepared samples of ZnO of good optical and electrical properties which can be used in many opto-electrical applications and mainly in the field of photovoltaic cells.

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