A Study of Structure and Optical Properties of ZnO Thin Films Deposited by Using Thermal Evaporation Technique under Different Flow Rate of Oxygen O_2

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Abstract

Zinc oxide (ZnO) transparent thin films with different oxygen flow rates (0.5, 1.0, and 1.5)Litter/min. were prepared by thermal evaporation technique on glass substrate at a temperature of 200℃ with rate (10±2)nm sec^{-1}, The crystallinity and structure of these films were analyzed by X-ray diffraction (XRD). It exhibits a polycrystalline hexagonal wurtzite structure and the preferred orientation along (002) plane. The Optical properties of ZnO were determined through the optical transmission method using ulta violet–Visible spectrophotometer with in wave length (300-1100)nm. The optical transmittance of the ZnO films increases from 75% to 85% with increase flow rate of O_2, and the optical band gap of ZnO films was found to be increased from (3.0) eV to (3.2) eV with increase flow rate of oxygen.The refractive index of ZnO flims increased from (1.1) to (1.27) with increase flow rate of O_2 .

Key word : ZnO thin films, X-ray diffraction, Optical properties.

Introduction

Zinc oxide (ZnO) is n – type semiconductor of wurtzite structure exhibits many interesting properties, such as a wide energy band gap (3.37)eV, high excitonic binding energy (60)meV, high photoconductivity and important piezoelectric and pyroelectric properties. These behaviors make this compound useful for numerous technological applications, such as transparent conducting electrodes, ultraviolet, surface acoustic waves filters, gas sensors and recently in nanoelectronics and photonics. A variety of techniques such as pulse laser deposition, molecular beam epitaxy, vapor phase transport, chemical vapor deposition, r.f. sputtering, spray pyrolysis, ect. [1-3]. However, not much attention has been paid to the preparation of ZnO films by thermal oxidation of metallic Zn thin films, such a method is a relatively simple and low- cost procedure that does not require any calalyst or higher- temperature growth[4]. Many of searchers works of ZnO thin films as [1-4]. The aim of this paper is study structural and optical properties at different flow rates of oxygen.

Experimental

The metallic Zn films were deposited by thermal evaporation under vacuum onto clean glass substrates by vacuum evaporation technique. A cylinder chamber of height 8 cm, closed at the top by the substrate holder. The residual pressure in the standard vacuum system used was about 4.5×10^{-5} Torr and heating glass substrates at 200℃ . These films prepared at thickness were 500 ± 50 nm with rate (10 ± 2)nm sec^{-1}.

After preparation Zn films were heated under 400℃ at different flow rates of O_2 (0.5, 1.0, and 1.5) litter/min. for one hour. The crystalline structure of the studied films was investigated by X-ray diffraction (XRD) analysis using Cu-K_α radiation (λ= 1.5418) A° in the rang $2\theta = 20^\circ$ -80°. The films transmittance was measured using a UV-VIS spectrometer in the wavelength range (300 – 1100) nm .

Results and Discussion

Fig. (1) shows the XRD spectra of ZnO films prepared at different flow of oxygen which are (0.5, 1.0, and 1.5) litter/min. , respectively. For all as – grown films, a strong peak corresponding to (002) plane at around 34°, which presented the ZnO films are polycrystalline and have hexagonal structure.

The diffraction intensiveities of the (002) (100) and (101) direction are increased with theincrease of flow rate oxygen, that means improving crystalline material[5,6].

Fig. (2) displays stress along the c-axis for the ZnO films as a function of the flow rate of oxygen. The variation of the curve presents that with oxygen flow rate increase, stress along c-axis slightly increases and the film stress σ along the c-axis is given by $(C_\circ - C)/C_\circ$ where C is the lattice constant obtained from the (002) direction in the XRD spectra and $C_\circ$ is 0.5205 nm measured from bulk ZnO[5].

The stress σ along the c- axis with a hexagonal crystal structure is given by the following equation [5]:-

$$\sigma = \left[\frac{2C_{13}^2 - C_{33}C_{11} - C_{33}C_{12}}{C_{13}}\right] \frac{C_\circ - C}{C_\circ} \ldots\ldots\ldots (1)$$

Where C_{ij} is elastic stiffness constant of ZnO, C_{11}=209.7 GPa, C_{12} 121.1=GPa, C_{33}= 210.9 GPa and C_{13}= 105.1 GPa [5]. Substituting these values in the above equation gives[5]:-

$$\sigma\left(N/m^2\right) = -453.6 \times 10^9 \frac{C_\circ - C}{C_\circ} \ldots\ldots\ldots(2)$$

According to the above equation, if the stress σ is positive, the biaxial stress is tensile, if the stress is negative, the biaxial stress is compressive [5]. All the as- grown films exhibit tensile stress, when the flow rate of oxygen increases from 0.5 to 1.5 litter/min., the tensile stress increases from 3.962×10^{6} N/m^2 to 4.092×10^6 N/m^2, and this result breffare from result reference [6].

Fig. (3) shows the transmission for ZnO thin films with wavelength at different oxygen gas flow, we can see from this Fig. the transmission increases with the increase of oxygen flow rate, and was attributed mainly to the improvement of crystalline structures of the films. In near infrared (NIR) spectrum region with (900-1100)nm, the average transmission is more than 85% for flow rate 1.5 litter/min. .

The optical absorption edge was analyzed by the following equation [7]:-

$$\propto hv = B\left(hv - E_g\right)^{0.5} \quad \ldots\ldots\ldots\ldots..(3)$$

Where B is a constant and $\propto$ is absorption coefficient. The variation of $(\propto hv)^2$ with photon energy (hv) for the ZnO thin film is shown in Fig. (4). It has been observed that plots of $(\propto hv)^2$ versus hv are linear over a wide range of photon energies indicating the direct type of transitions [7]. The optical band gap of ZnO films increases from (3.0 to 3.2) eV with the increase of oxygen gas flow rate because of increase crystallinity material and decreased crystalline defect, so position levels reduced near valance and conduction bands.

The refractive index is an important parameter for optical materials and application, Thus, it is important to determine optical constants of the films. The refractive index of the films was determined from the following relation [8]:-

$$n = \frac{1+R}{1-R} + \sqrt{\frac{4R}{(1-R)^2} - K^2} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots..(4)$$

Where R is the reflection, and K is the extinction coefficient.
The extinction coefficient can be obtained from the experimental expression [7]:-

$$K = \frac{\propto\lambda}{4\pi} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots(5)$$

Where λ is the wave length. from Fig.(5), the films show that the refractive index values decrease with the increase of flow rate oxygen. The extinction coefficient dependence on wavelength is shown in Fig.(6), where it is decreased with the increase of flow rate oxygen.

Conclusions

We have deposited ZnO thin films on glass substrate using thermal evaporation technique at different oxygen flow rates at 200 ℃ . The XRD measurement showed the intensity of ZnO films increase with the increase of oxygen gas flow. The tensile stress along the c-axis in the as – grown thin film attributed to the common effect of amorphous substrate and extruding between grains. The optical transmittance is 80% at flow 1.5 Litter/min., and a value of about 3.2 eV for the optical band gap at flow rate 1.5 Litter/min. . These behaviors are comparable with those of ZnO films grown by other preparation techniques, and it may be considered that the deposited crystalline ZnO thin film was suitable for many optical devices, such as solar cells and electrodes.

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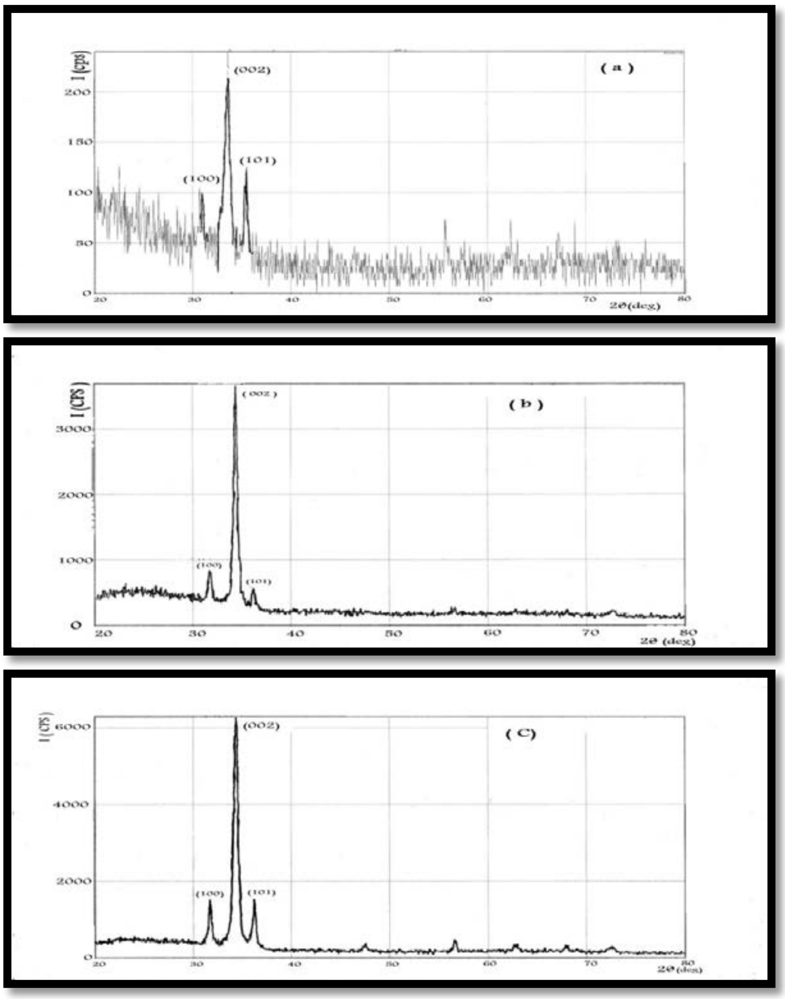

Fig. (1): XRD patterns of ZnO thin films at different flow rates of O_2: (a) 0.5, (b) 1.0 and (c) 1.5 litter /min

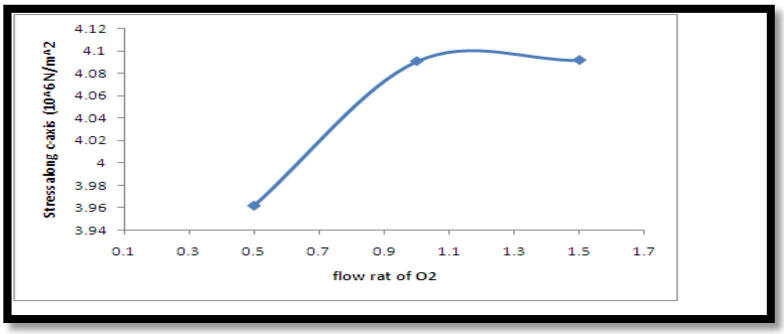

Fig.(2): Stress along the c-axis for ZnO films as a function of the flow rate of O_2

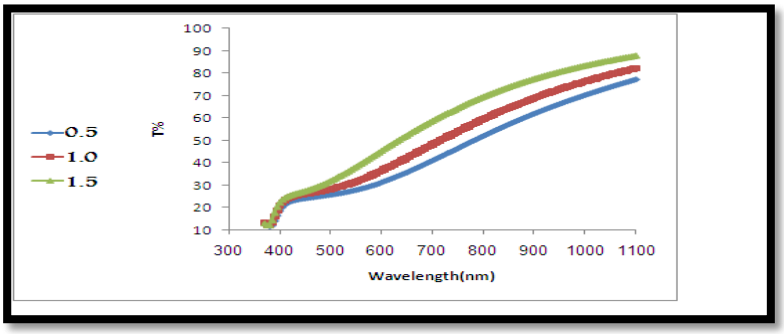

Fig.(3): Transmission spectra for ZnO films different flow rat of oxygen

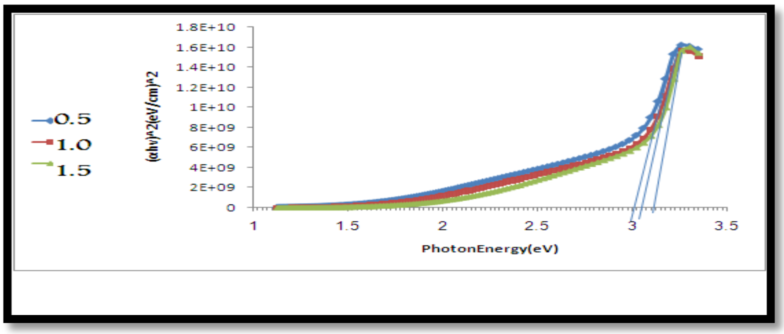

Fig.(4): Optical energy for ZnO films at different flow rat of oxygen

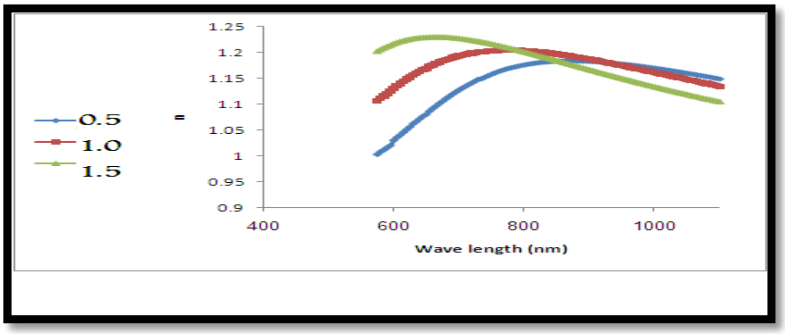

Fig.(5): Refractive index with wave length for ZnO films at different flow rate of oxygen.

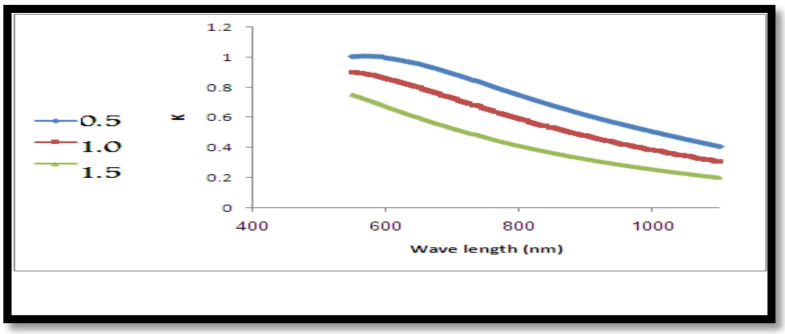

Fig. (6): Extinction coefficient with wave length for ZnO films at different flow rate of oxygen.

دراسة الخصائص التركيبية والبصرية لاغشية اوكسيد الخارصين الرقيقة المرسبة بطريقة التبخير الحراري الفراغي عند اختلاف معدل تدفق الاوكسجين (O_2)

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الخلاصة
حضرت اغشية رقيقة لاوكسيد الخارصين (ZnO) بتقنية التبخير الحراري الفراغي على قواعد من الزجاج المرسبة بدرجة حرارة ا ساس 200°C وبمعدل ترسيب $nm\ sec^{-1}$ (10 ±2)بمعدل تدفق مختلف لغاز الاوكسجين (0.5, 1.0,and 1.5) litter/min.)، ان حيود الاشعة السينية بينت ان الغشاء ذو تركيب سداسي متعدد التبلور والاتجاه السائد هو للمستوي (002) . حددت الخواص البصرية لاوكسيد الخارصين عن طريق النفاذية البصرية باستعمال مطياف ZnO ultra violet – visible لمدى الاطوال الموجية (300-1100)nm فالنفاذية البصرية تتغير من 75% الى 85% بزيادة معدل تدفق الاوكسجين وفجوة الطاقة البصرية للاغشية تتراوح بين (3.0)eV الى eV (3.2) بزيادة معدل تدفق الاوكسجين. معامل الانكسار لأغشية ZnO تزداد من 1.1 الى 1.7 بزيادة معدل تدفق الاوكسجين.

الكلمات المفتاحية : اغشية ZnO الرقيقة، حيود الاشعة السينية، الخصائص البصرية.