Effect of Al-doping on the Optical Properties of ZnO Thin Film Prepared by Thermal Evaporation Technique

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Abstract. Aluminum doped ZnO (AZO) thin films doped has been prepared by thermal evaporation method onto glass substrates. The optical characterization has been studied by UV-Visible Spectrophotometer in the range (300-1100) nm. The transmittance increased with increasing Al-doping, while absorption decreases with increasing Al-doping content. The energy gap (E_g) values increased with increasing Al-doped in the ZnO:Al thin films. The refractive index, extinction coefficient, and real and imaginary dielectric constants obtained results show that doping content strongly affects the optical parameters of AZO thin films. Optical constants tend to decrease with increasing doping content.

Introduction

ZnO is an II-VI transparent conducting oxide (TCO) touted as a material of choice for short-wavelength optoelectronics [1-2]. This semiconductor has several favorable properties: good transparency, high electron mobility, wide and direct band gap of 3.37 eV at 300 K [3]. ZnO is an n-type semiconductor and its conductivity can be controlled by thermal treatment or by adequate doping [4]. ZnO thin films have attracted considerable interest in recent years for application such as: transparent conducting material in liquid crystal displays [5], solar cell [6], gas sensors [7], and catalyzers [8]. Various techniques such as molecular beam epitaxy (MBE) [9], pulsed laser deposition (PLD) [10], magnetron sputtering [11], chemical vapor deposition (CVD) [12], atomic layer deposition [13], electron beam evaporation [14], hydrothermal method [15], and sol-gel process [16] have been applied to ZnO thin film preparation.

Many researches of Al-doped ZnO thin films are presented. The Al-doped ZnO thin film showed high transmittance and crystallinity [17].

In this work, it study the optical properties of ZnO:Al thin films prepared by thermal evaporation method.

Experimental Techniques

Zinc Oxide and Aluminum (99.99% purity) were mixed at varying doping percentages of Aluminum (0, 0.002, 0.004, 0.006) wt.%. Glass substrate was cleaned by distilled water and ethanol to remove surface contaminants and allowed to dry completely. The substrate was then mounted on a rotating substrate holder and the compound was then placed in a Molybdenum boat in Thermal evaporation system. The chamber was covered tightly and pumped down to 1×10^{-7} mbar. Thickness of the thin films has been measured by the optical thin film measurement and found to be 125 nm. The optical properties have been studied by UV-Visible Spectrophotometer in the rang (300-1100) nm.

Results and discussion

The optical properties of pure ZnO and have doped with different content of Al (0, 0.002, 0.004, 0.006) wt.% at thickness 125 nm with annealing temperature at 573 K were investigated using UV-Visible Spectrophotometer in the range (300-1100) nm.

In general, we can observe that the transmittance increase with increasing wavelength for all prepared thin films. Fig. 1 shows the transmittance spectra of pure and Al-doped ZnO thin films with different content of Al. This result may be due to the reduction of the voids in the sample and the improvement of the homogeneous structure with uniformly distributed particles, thereby increasing the optical scattering. The increased optical transmittance is related to the crystallinity of the film, which is in accordance with the findings of workers Kim H. et al. [18] and E. M. Mkawi et al. [17].

Fig. 1: Transmittance spectra as a function of wavelength of ZnO:Al thin films.

Fig. 2 shows the absorbance spectra of pure and Al-doped ZnO with different content of Al. From this figure, it can observe that the absorbance decreased with increasing Al-doped ZnO thin films.

Fig. 2: The absorbance spectra as a function of wavelength of ZnO:Al thin films

The absorption coefficient (α) of ZnO:Al thin films system has calculated by using equation [19]:

$$\alpha = 2.303 \frac{A}{t} \quad (1)$$

In general, we can observe that the absorption coefficient decreased with decreasing wavelength for all prepared thin films as shown in Fig. 3 that represents the relationship between

the absorption coefficient spectra of pure and doped ZnO with different content of Al and wavelength. This result is agrees with the findings of other workers Mujdat C. *et. al* [20].

Fig. 3: The absorption coefficient spectra as a function of wavelength for ZnO:Al thin films.

The optical energy gap of ZnO:Al thin films system has determined by using equation [21]:

$$\alpha h\nu \approx [h\nu - E_g]^{1/2} \quad (2)$$

The plot of $(\alpha h\nu)^2$ with energy ($h\nu$) indicates that ZnO films are direct transition type semiconductors at all thin films. The photon energy at the point where $(\alpha h\nu)^2$ is zero is E_g .

From the Fig. 4, shows the energy gaps of pure and Al-doped ZnO thin films with (0.002, 0.004, and 0.006)wt.% aluminum. An obvious increase is observed for the values of the energy gap with the increase of the Al content. This increase is explained by the preposition that the ZnO:Al films are semiconductors in which the Fermi level lies in the conducive band which means that the levels at the bottom of the conductivity band are occupied by electrons and the shielding of electronic traveling to these levels. These results show a good agreement with the findings of research Youssef A. *et. al* [22]. These results are listed in Table 1.

Fig. 4: A plots of $(\alpha h\nu)^2$ verses photon energy ($h\nu$) of ZnO: Al thin films.

Table 1: The optical energy gap of Al-doping ZnO thin films.

Type of thin film	Optical energy gap (eV)
ZnO	3.5
ZnO:0.002%Al	3.65
ZnO: 0.004% Al	3.75
ZnO: 0.006% Al	3.85

The refractive index (n) of ZnO:Al thin films system has determined by using equation [23]:

$$n = \left(\frac{4R}{(R-1)^2} - k^2 \right)^{1/2} + \frac{(R+1)}{(R-1)} \quad (3)$$

The behavior of the extinction coefficient of pure and Al-doped ZnO thin films with different content of Al. The variation of the refractive index versus wavelength in the range (300-1100) nm were shown in Fig. 5. It can notice from the figure that the refractive index, in general, decreases with the increasing of Al content may be mainly attributed to an increase of the carrier content in the ZnO:Al films. For all the films, this is in agreement with S. Bandyopadhyay *et al.* [24] and Xue *et al.* [25].

Fig. 5: Variation of refractive index with wavelength for pure and Al-doped ZnO thin films.

The extinction coefficient (k_o) for ZnO:Al thin films system has determined by using equation [26]:

$$k_o = \frac{\alpha\lambda}{4\pi} \quad (4)$$

In general, we can observe that the extinction coefficient decrease with increasing of Al content for all prepared thin films. Fig. 6 shows the extinction coefficient of pure and Al-doped ZnO with different content of Al. This is in agreement with Min S. K. *et al* [27].

Fig. 6: Variation of extinction coefficient with wavelength of ZnO:Al thin films.

The real and imaginary dielectric constants (ϵ_r and ϵ_i) of ZnO:Al thin films system have determined by using equations [26]:

$$\epsilon_r = n^2 - k^2 \quad (5)$$

$$\epsilon_i = 2nk \quad (6)$$

The variation of real (ϵ_r) and imaginary (ϵ_i) parts of dielectric constant values versus wavelength for pure and Al-doped ZnO with different Al content. In general, It can observe that the ϵ_r and ϵ_i decrease with increasing of Al content for all prepared thin films. Figs.(7-8) shows the ϵ_r and ϵ_i of pure and Al-doped ZnO with different content of Al. This is in agreement with Mujdat C. *et. al* [20].

Fig. 7: Variation of ϵ_r with wavelength of ZnO:Al thin films.

Fig. 8: Variation of ϵ_i with wavelength of ZnO:Al thin films.

Conclusions

- 1- The transmittance spectra increased with the increasing of Al-doped ZnO thin film, the highest transmission is (92%) , while the absorption spectra decrease with increasing Al-doping. These films more suitable for solar cell fabrication.
- 2- The value of the band gap is enhanced from 3.5 eV for un-doped ZnO to 3.85 eV for ZnO:0.006%Al.
- 3- The absorption coefficient, refractive index, extinction coefficient, and the real and imaginary parts of dielectric constant are obtained results show that doping content strongly affects the optical parameters of AZO thin films. Optical constants tend to decrease with increasing doping content.

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