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Structural and optical properties of ZnO:Al films prepared by the sol–gel method

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Abstract

Aluminum doped zinc oxide polycrystalline thin films (ZAO) were prepared by sol-gel dip-coating process. The sol was prepared from an alcoholic solution of zinc acetate using DEA (Di Ethyl Amin) as a stabilizer. The structural characteristics have been studied by X-ray diffractometry and UV–Vis-NIR spectroscopy. The films are transparent from the near ultraviolet to the near infrared, presenting an absorption cut-off at almost 290 nm. An optical band-gap of 3.32 eV have been measured from the transmittance spectroscopy experiments when the quantity of aluminum in the sol was varied from 0% to 4%.

The best films were obtained for the ZnO films containing 2% of aluminum prepared at 550°C by adding $Al(NO_3)_3.9H_2O$, with the best transmittance over the range 400-1000 nm, a change in the E_g value from 3.16 eV to 3.18 eV when the ratio of Al was changed from 0% to 2%, with best structural orientation at (100) (002) (101). The smoothest surface was also obtained by SEM for a dopant 2% value of the Aluminium.

Author Keywords: Sol–gel method; ZnO:Al; ZAO, SEM

1. INTRODUCTION

ZnO thin films have interested as transparent conductor, because the n-type ZnO thin film has a wide band-gap semiconductor (E_g=3.2 eV), and high transmission in the visible range, and ZnO thin films can take place of SnO_2 and ITO because of their electrical and optical properties and its excellent stability which has been mentioned widely.[1][2]

ZnO thin films have been prepared with different processes such as pulsed-laser deposition, chemical vapor deposition spray pyrolysis and sol–gel process etc. Among them, the sol–gel technique[3] offers the possibility of preparing a small coating as well as a large-area coating of ZnO thin films at low cost for technological applications.

In this work, we deal with structural and optical properties of ZnO:Al thin films prepared by the sol–gel process.[4][5], in order to increase the band-gap and the optical transmittance.

2. EXPERIMENTAL PROCEDURE

ZnO thin films were prepared by the sol–gel method. As a starting material, zinc acetate dehydrate was used. We put 0.1 mol/100ml from $Zn(CH_3COO)_2.2H_2O$ in flask 100 ml and complete it to the mark with propanol-2 then stirred it for fifty minutes until we get a milky liquid, and we added drops from DEA (Di Ethyl Amin) as a stabilizer to get a transparent liquid. It has been leaved in lab's temperature about 24 hour, then we filter it , and we obtained the Sol ready to be used to prepare the film by spinner. The process begin with heating the film in the furnace at 550 °C for one hour, then at 200 °C for 15 minutes to obtained the crystalline form of the ZnO film [6][7].

In order to study the effect of aluminum concentration on the structural and optical properties of Al-doped ZnO (ZnO:Al) thin films, four values of dopant concentration were prepared (1, 2, 3, 4 wt. %) by adding Al (NO3)3.9H2O to the starting material.

The XRD spectrums of the samples were recorded at room temperature using a Phillips PW 1480 Albaath university labs. The thickness of the thin films was measured using a VEECO DEKTAK3 profilometer. The surface morphology of the films was analyzed using a scanning electron microscope (SEM). The optical transmittance was measured using a UV–VIS–NIR Cary 50 Scan spectrophotometer in the wavelength range from 400 to 1000 nm in Montpellier II University laboratories.

3. RESULTS AND DISCUSSION

3.1 Crystalline characterization by XRD measurements

Figure 1 displays the XRD spectrum of ZnO films. Three lines (100) at 2θ = 32°, (002) at 2θ = 34°, (101) at 2θ = 37° are pointed, they will be considered for structural characterization of ZnO.

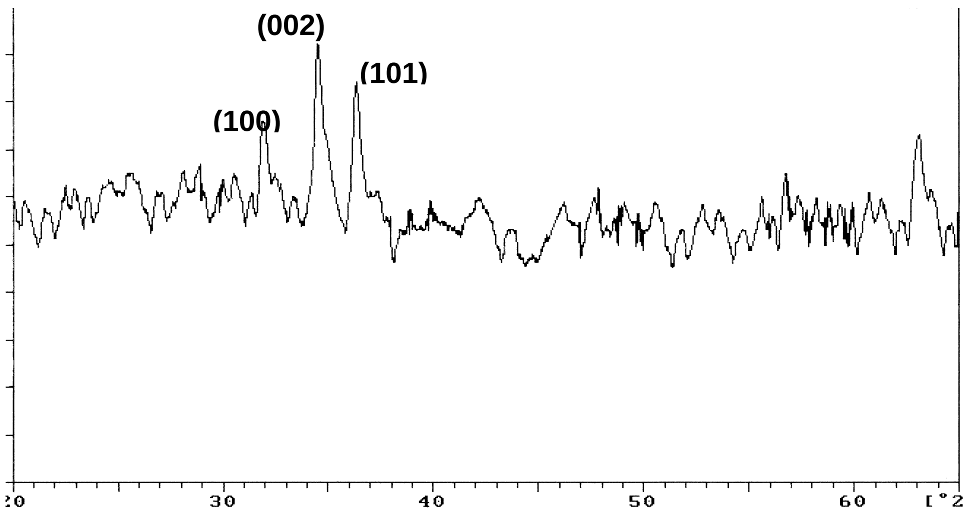

Figure 1: ZnO thin film XRD Spectrum.

We measured the XRD spectra for ZnO:Al with different Al ratio (weight) from 1% to 4 % and found the following results represented in figure 2.

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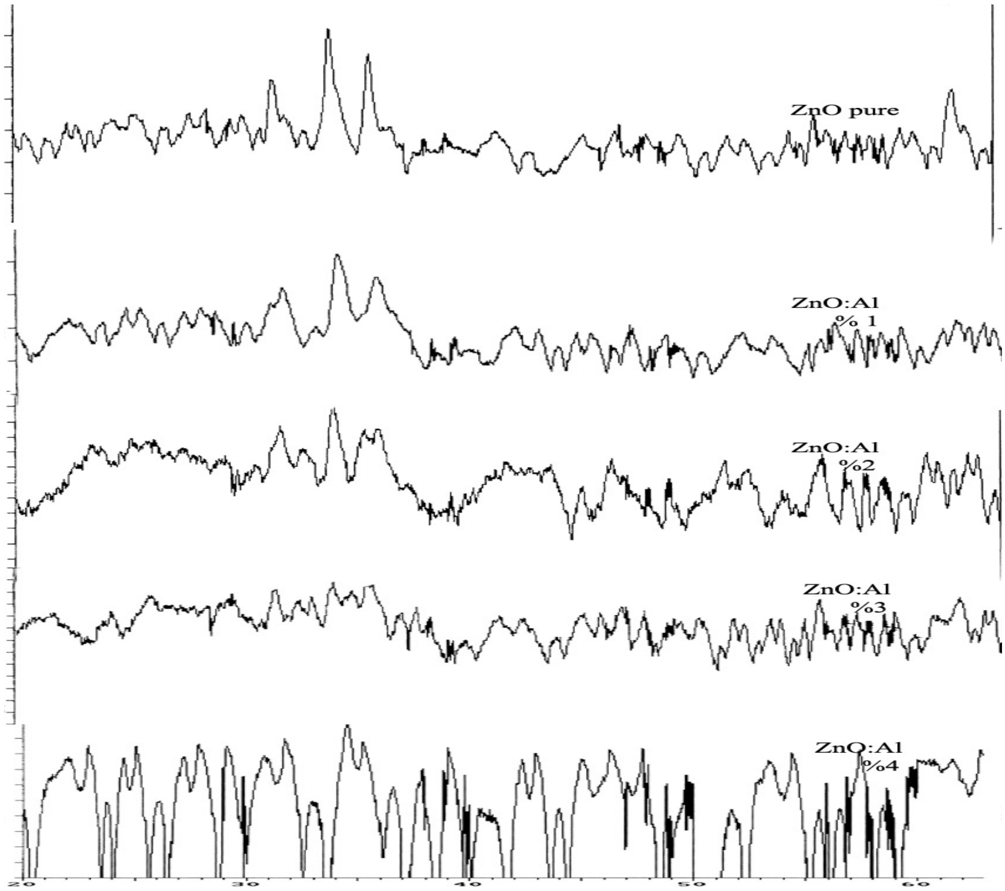


Figure 2: XRD Spectrums for ZnO:Al thin film. Al concentration (in weight) are indicated?

A significant evolution of the resolutions of ZnO (100), (002) and (101) characteristic lines is obtained. The increase of Al concentration in the starting material for the ZnO preparing process, leads to an amorphous stage of the films which appears at concentrations higher than 2%.

3.2 Characterization of surface and thickness of ZnO:Al films

We took SEM pictures for ZnO prepared by spin coating then we measure the thickness by SEM and Pro-filometer. Results are given in figure 3 and table 1 for ZnO films.

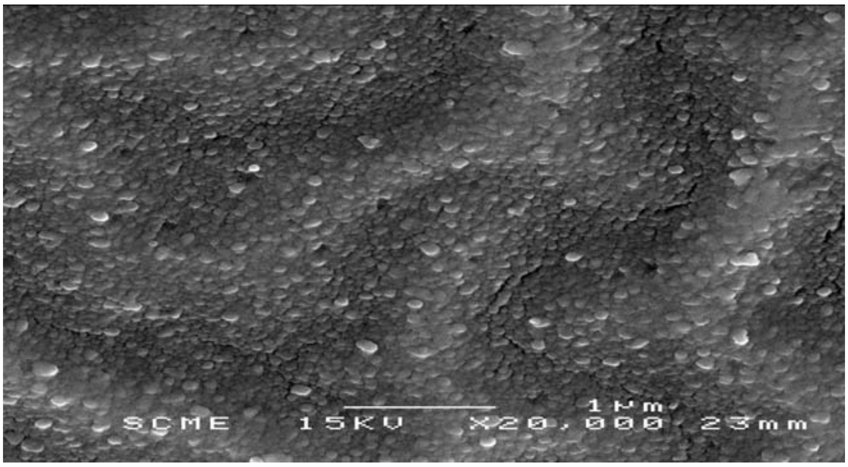
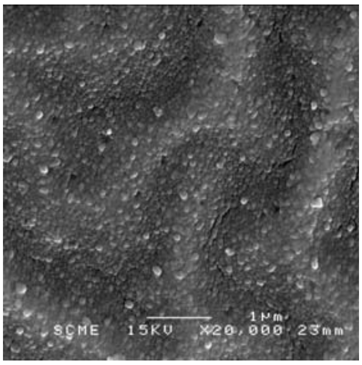


Figure 3: ZnO thin film observed by scanning electron microscope.

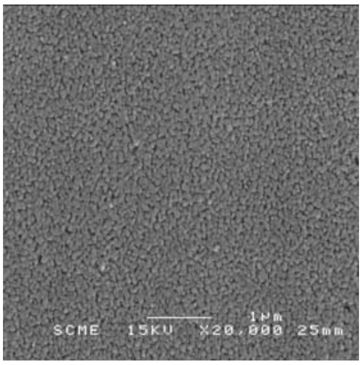
Film type	SEM	Profilometer
ZnO / Spin coating	850 nm	920 nm

Table 1: Thickness of ZnO films obtained by SEM and Profilometer.:

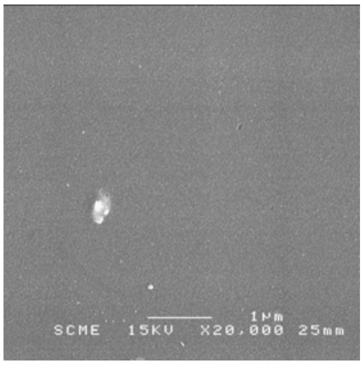
Figure 4 displays the SEM observations for ZnO:Al films.



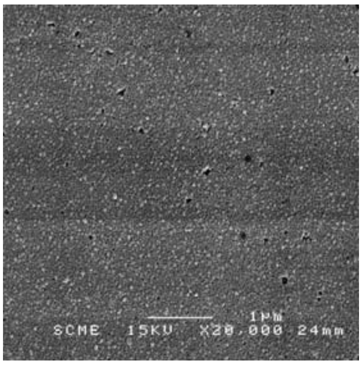
SEM for ZnO pure



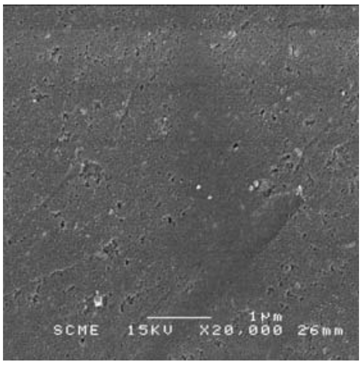
SEM for ZnO:Al 1%



SEM for ZnO:Al 2%



SEM for ZnO:Al 3%



SEM for ZnO:Al 4%

Figure 4: SEM pictures of ZnO:Al thin films obtained with incorporation of 1-4% (wt) Al.)

The important results concern the smoothest surface (grain size) for a 2% (wt) Al impurities in ZnO:Al films, together with a first decrease of microstructures when increasing Al impurities until 2%.

3.3 Optical properties

Figure 5 displays the measured transmittance of the ZnO and ZnO:Al films in the rage 400nm to 1000 nm.

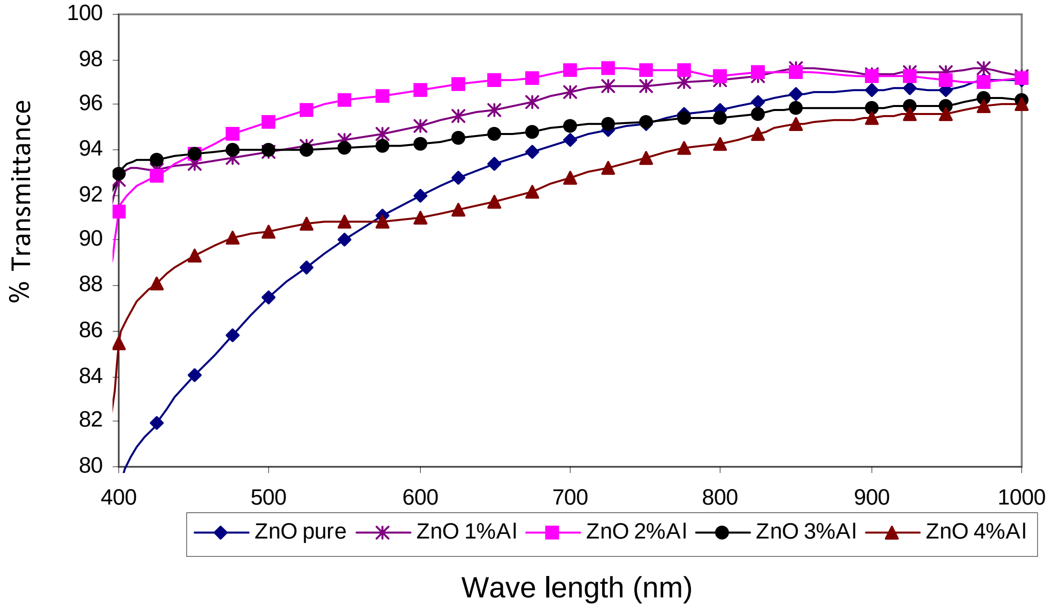
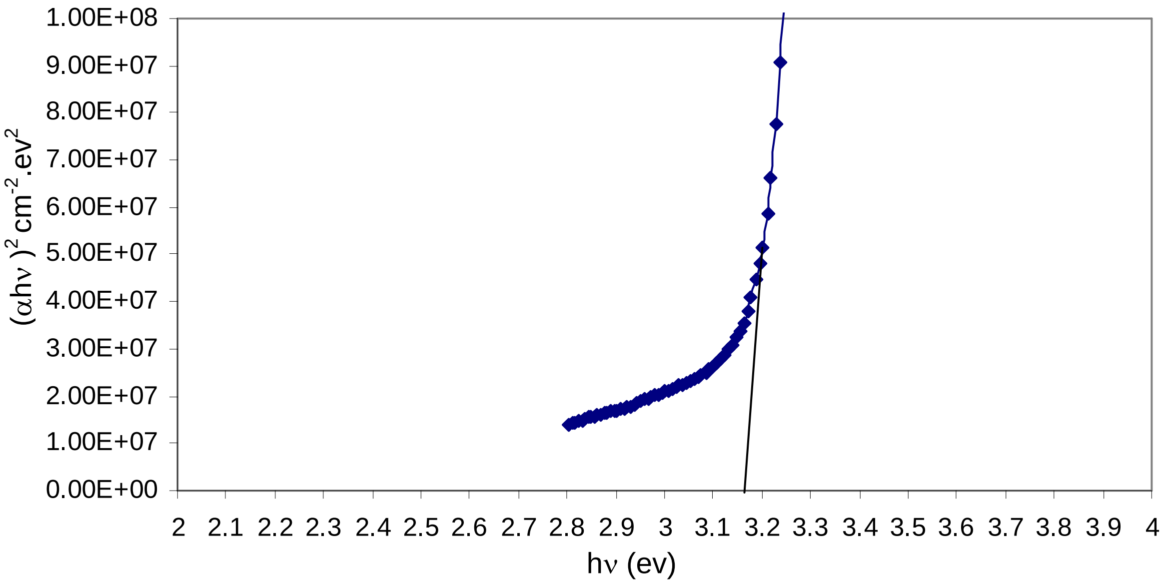


Figure 5: ZnO and ZnO:Al(1-4%) thin films transmittance spectroscopy

Results show that the introduction of 2% of Aluminium in Zn0 films increases the film transmittance for lower values of the wavelength in the range 400 nm to 1000 nm and enable to obtain a variation of the transmittance lower than 7% in this range.

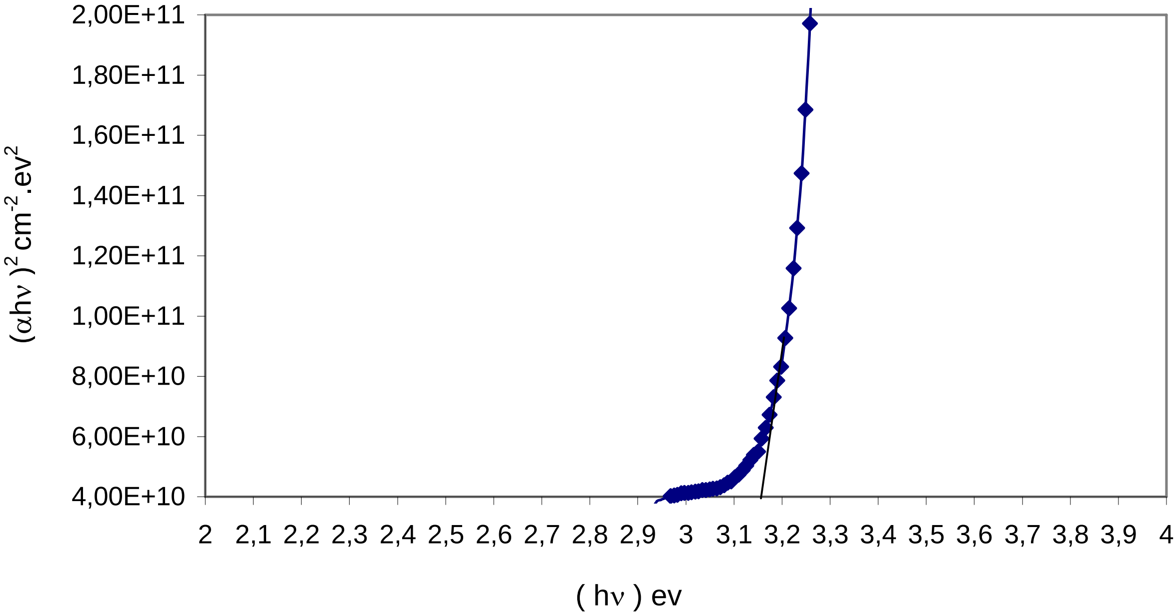
3.4 Energy band gap

The energy band gap could be obtained from spectroscopy measurements /6/. We found the following results:

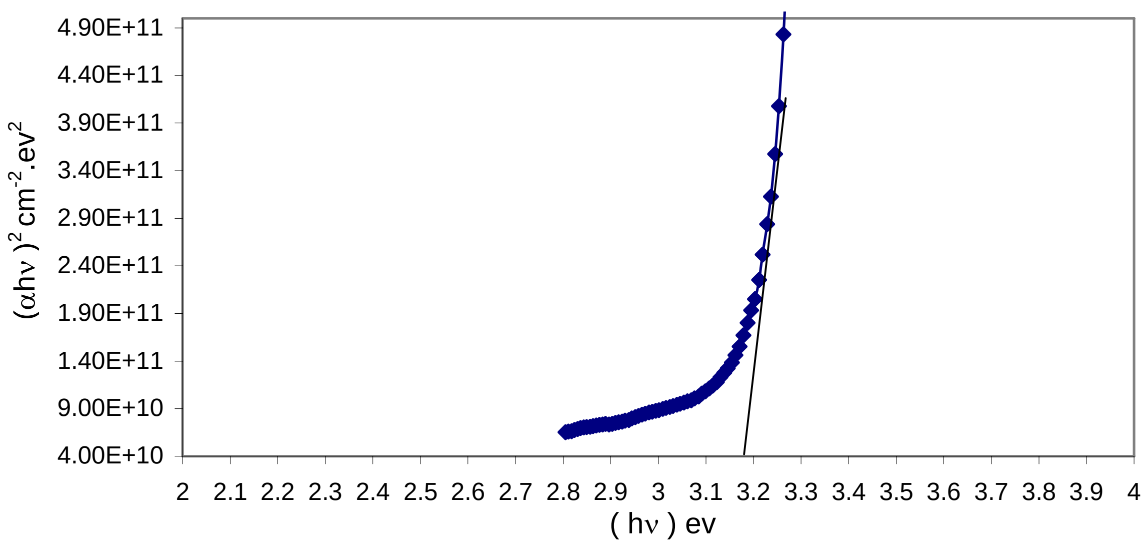


Fig(6): Measurement of energy band gap for ZnO pure thin film

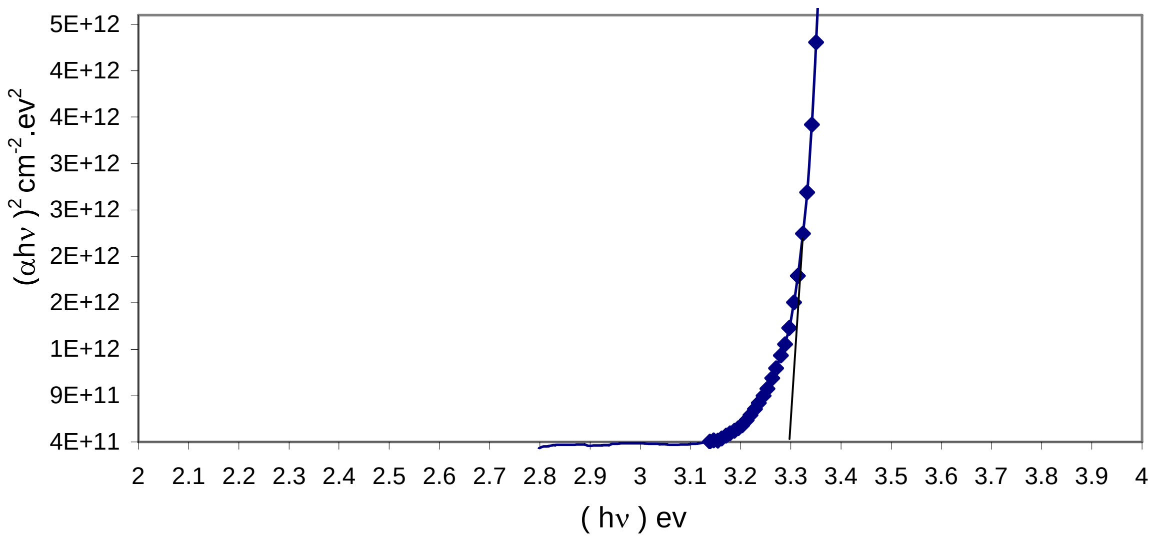
In order to study the effect of Al concentration on the energy band gap values of ZnO:Al thin film, we added Aluminum as a dopant to the sol by using Al $(NO)_3.9H_2O$ (1-4 %). Figure 7 to10 display $(\alpha h\nu)^2$ as a function of $h\nu$ [1][6], α is the absorbent coefficient of the thin film.



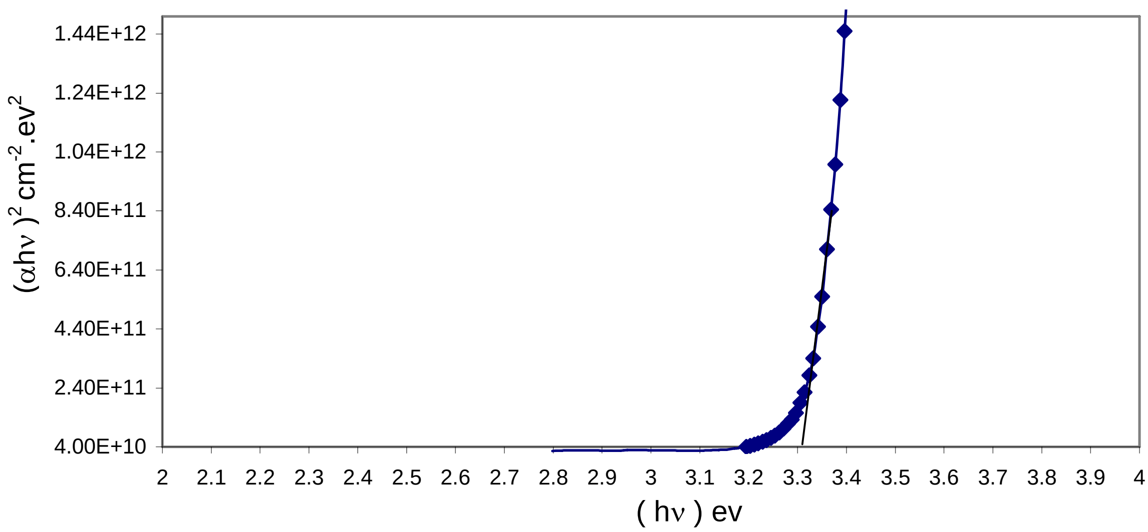
Fig(7): Measurement of energy band gap for ZnO:Al 1% thin film



Fig(8): Measurement of energy band gap for ZnO:Al 2% thin film



Fig(9): Measurement of energy band gap for ZnO:Al 3% thin film



Fig(10): Measurement of energy band gap for ZnO:Al 4% thin film

Table 2 gives the evolution of the band gap. The increase of is significant for doping Al values larger than 2%.

Type of the film	Energy band gap value (eV)
ZnO pure	3.16
ZnO:Al 1%	3.16
ZnO:Al 2%	3.18
ZnO:Al 3%	3.30
ZnO:Al 4%	3.32

Table 2: The energy band gap obtained by spectroscopy measurements.

4. CONCLUSIONS

ZnO thin films were prepared on glass substrates by spin-coating method, and the effect of Al ratio as a dopant of the films were studied by measuring structural and optical properties. The films which were having a 2% Al ration showed the preferred (1 0 0), (0 0 2), (1 0 1) orientations of ZnO:Al. SEM images of the films showed a smooth grain size and the microstructures of films became denser. The optical transmittance was between 91% and 98% in the visible and near IR regions. An increase of E_g values from 3.16 eV to 3.32 eV was obtained when the ratio of Al changes from 0% to 4%.

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