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Investigation of aluminuim doping on structural and optical characteristics of spray pyrolysis assisted dip coated zinc oxide films

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ZnO film is prepared by dip coating on a glass substrate. Al/ZnO film is prepared by spray pyrolysis method on a glass substrate. Studying the structure of Al-doped 4% ZnO powder, and its optical properties, where XRD patterns of Al/ZnO powder after heat treated at 430 °C show main sharp peaks, the higher intensity (refer to the planes (100); (002); (010); (102) and (110); it indicates the formation of hexagonal wurtzite structure with more crystallites. It is found that the best percentage (%) for doping zinc oxide with aluminum is 4%, showed better performance of the high optical transmission; ascribable to regularity; little surface bumpiness of thin films, and found that the result for the transmittance of the films decreases with the increase in Aluminum doped by more than 10%.

Keywords: Structural and optical properties, dip coating, spray Pyrolysis.

1. INTRODUCTION

ZnO and doped films are obtained by soft chemistry starting with zinc acetate dihydrate. and Al(III) isopropoxide in absolute ethyl alcohol. The membranes are deposited by dip coating technology on a silicone substrate and thermally treated at 500 ° C for one hour. According to elliptical spectral data, more porous and thinner films are obtained, with a smaller refractive index in the case of Al/doped ZnO films compared to ZnO films.

Porous and thinner films, with smaller refractive index are obtained in the case of Al /doped ZnO films as compared with ZnO films. Both ZnO and Al-doped ZnO films presented high electrical resistivity [1]. One popular and well-respected method for treating thin films is spray deposition. Chemical vapor deposition has proven to be an effective method for precipitating several types of oxides [1-5]. This method's two primary uses are for deposition on a vast surface and operating at atmospheric pressure.

Because air is used instead of a vacuum system, as is typically the case with many sedimentation processes (sputtering, CVD, and evaporation), the approach is incredibly easy.

Spray precipitation works by mechanically converting a solution into a stream of droplets through the use of ultrasound or compressed gas. After being sprayed, the droplets react to form a solid coating on a heated surface. Subsequently, the primary factors governing the spray sedimentation processes and the characteristics of the deposited membranes are the properties of the solution employed and the façade / substrate drop thermodynamics [6-9].

2. EXPERIMENTAL WORK

2.1. Materials

The materials for the synthesis the ZnO film are zinc acetate $Zn(CH_3COO)_2.{}_2H_2O$ from BDH /England and chromic acid H_2CrO_4 from CDH/India. Also; the materials used to prepare (AL/ZnO) are acetate of Zinc $[Zn(CH_3COO]_2.2H_2O$; acetate of Aluminum $Al(CH_3COO)_2.4H_2O$, Isopropyl alcohol $CH_3CH_2CH_2OH$, Acetic acid all from BDH /England, Methanol from Fluka company; and Acetone CH_3COCH_3 from Merck-France. Instruments used the X-ray diffractometer (XRD), and UV-vis spectrophotometer.

2.2. Procedure

2.2.1. Preparation of ZnO Film by Dip Coating Technique Metallurgy Method

Zinc oxide solution intended to put 0.1M of Zinc acetate (Zn $(CH_3COO)_2$ in DW and then mixed by using magnetic stirrer for 1 hr at room temperature. The tested pH of this solution is in the range of (8 – 9). This solution is taken in a beaker of (50ml) in volume. Hot distilled water is prepared at temperature in the range (90 to 95) °C. The class substrates must be cleaned with chromic acid; then by distilled water. The clean glass substrate is dipped in a solution of Zinc acetate for a few fractions of seconds and then immersed immediately in hot distilled water, this procedure is carried out 50 times in a row. The coated films are utilized to evaluate the structural and optical properties of ZnO films after being heated to 150°C for an hour in an oven with hot air [10].

2.2.2. Preparation Al Functionalized ZnO Films

Mixing 3: 1 isopropyl alcohol with deionized water; dissolve of 0.1 M acetate of zinc to the blend. The purpose of adding a modest amount of acetic acid is to make zinc acetate more soluble.; the solution is prepared in a beaker of a volume of (50 ml). In the ending-aluminum acetate (Al $(CH_3COO)_2.4H_2O$ is added to the solution with different molar concentrations (1; 2; 4; 10) [11].

The spray pyrolysis technique is employed to apply (Al/ZnO) thin films on a glass substrate. Glass substrates are cleaned with (methanol and acetone) using the ultrasonic technique for 5 minutes. Substrates are immersed in distilled water have a small amount of nitric acid (HNO_3) with a pH equal to" 5", and then dried up the glass substrates in the oven at room temperature for (15) minutes. The solution is placed in the glass nozzle. The deposited Al/ ZnO is heat treated by heat substrate in the range (420 to 430) °C. N_2 gas pressure (1-1.1 bar) is used to spray the solution; the diameter of the nozzle is 30µm, and the distance between the hot glass substrate and spray is kept around 10cm. The spray process is repeated '5' times continuously every (10) seconds to gate a homogenous surface [11]. Finely the structure properties; absorbance, and transmittance of (Al/ZnO) films are examined. To study the structural properties of Al/ZnO, a solution of (Al/ ZnO) is put in a beaker; and then post cure in the oven at a temperature of 430 C° with a flow of N_2 gas for '1' hour then milled by mortar hand to gate a fine powder; also study the absorbance and transmittance of (Al/ZnO) films are prepared.

3. RESULTS AND DISCUSSION

A uniform structure and compact structure, smoothing surface to get good adhesion with the glass substrate so that a film of Al-doped 4% ZnO; is chosen after annealed at (430C°) to get crystal structure. To take a peek at the powdered (Al/ZnO) structural properties. The powder is heat treated at 430 °C under nitrogen gas; the properties of Al-doped 4% ZnO powder annealed in the oven at a temperature of (430C°) for one hour in an N2 atmosphere; are observed in table 1. The examination of (X-ray) diffractometry is shown in Fig .1 with main standard peaks in the angle range from (20 to 60) degrees. The observed peaks can be seen in plans in the directions (100) ; (002) ; (010) ; (102) ; and (110), with high intensity and crystal size in the range (26.78 to 30.71)nm, which indicates the formation of hexagonal structure of (Al/ZnO), this agrees with [4].

Table 1 Structural properties of Al-doped (4%) ZnO powder heat treated at (430°C) in nitrogen gas for 1 hour.

(hkl)	2Theta (degree)	Intensity (a.u)	d-Spacing (A°)	FWHM (degree)	Crystal size (nm)
(100)	31.795	1153	2.812	0.305	28.3
(002)	34.446	918	2.601	0.283	30.71
(010)	36.273	1860	2.474	0.315	27.73
(102)	47.566	320	1.910	0.395	22.96
(110)	56.620	622	1.624	0.352	26.78

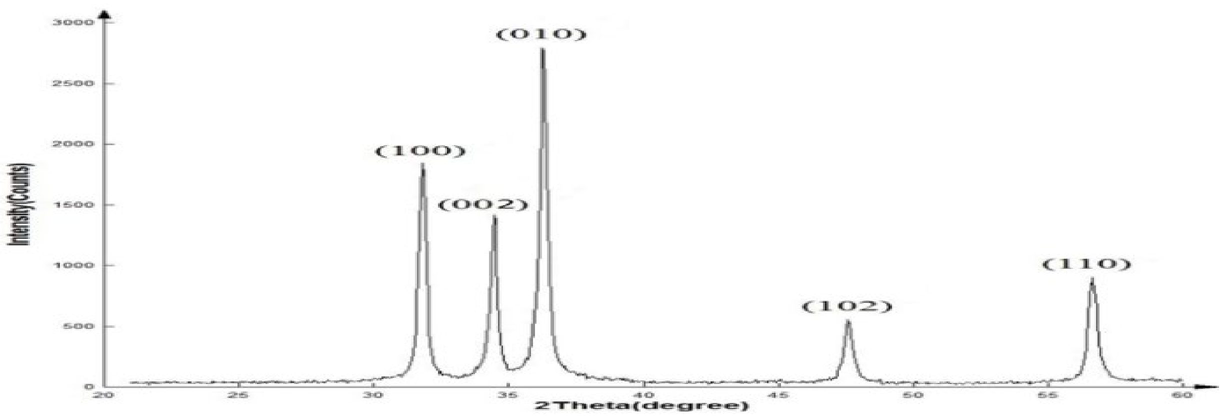

Figure 1 X-ray diagrams of Al-doped (4%) ZnO powder heat treated at 430 °C for one hour in an (N_2) atmosphere.

The spectra of UV-visible absorption and transmittance of Al/doped ZnO, (1%; 2%; and 4%) thin films are shown in Fig 2. and Fig 3. deposited on a glass substrate. The absorption spectra; in Fig2. display the percentage-based absorbance behavior for both pure (ZnO) and Al/doped ZnO. (1%; 2 %, 4%) wt% film's measured wavelength between (310 and 810) nm. The edge of the absorption Al/doped (ZnO) (1%) films rise with the shift at lower wavelengths and become's less sharp at a higher wavelength. The edge for absorption of Al-doped ZnO (2; 4 %) films is low at low wavelength. Al-doped ZnO (10 %) films are higher at low wavelengths these results agree with [12] and [13]. The values of transmissions of the films are low at short wavelengths and became high values at longer wavelengths; so, the films behaved as a transparent specimen at long wavelengths. The transmittance curve of the Al/doped ZnO

(1%) within easy reach of 100% of transmittance, and (2%) films are less than 100%. The films' transmittance first increased as Al doping is increased up to 4%; with a shift in optical band edge observed; and transmittance increases more than (100%) in the visible region; this improvement might be attributable to higher of vertical alignment, low surface roughness, and uniformity distribution of the film.

The improvement in the transmittance and shifting in the optical spectrum of Al/doped ZnO thin films may be due to the phase in thin films, the optical transmittance of the film's decrease with the increase in Al/doped more than 10%; this may be due to degradation of the films and these results agree with [12]; and [13]. The results of energy gup for Al-doped ZnO, (1%,2%,4%, and 10%), received in Fig 4.; values greater than 3eV with increasing aluminum concentrations; The extra carriers that the aluminum atoms supply is what cause and facilitate the Fermi level's shift towards the conduction band. Band gap rises as a result.

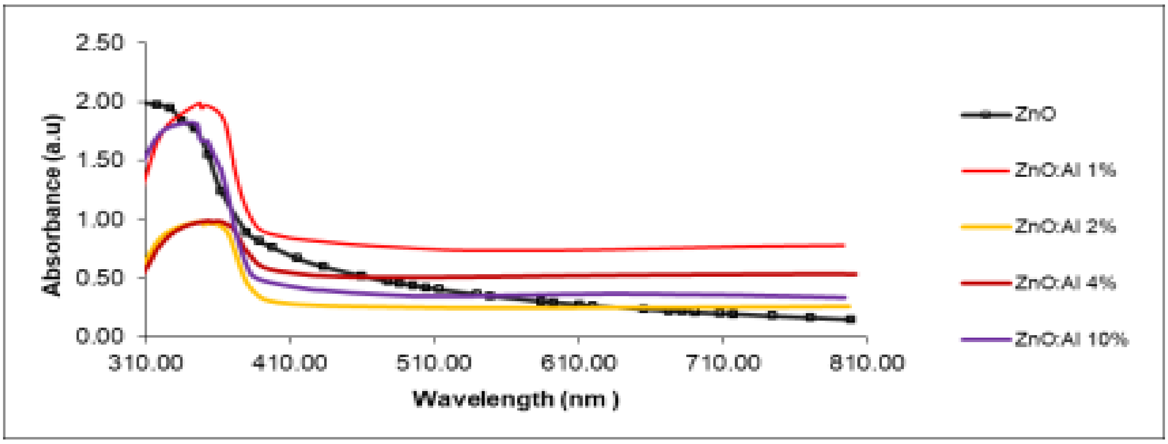

Figure 2 Absorbance behavior of ZnO and Al doped ZnO thin films.

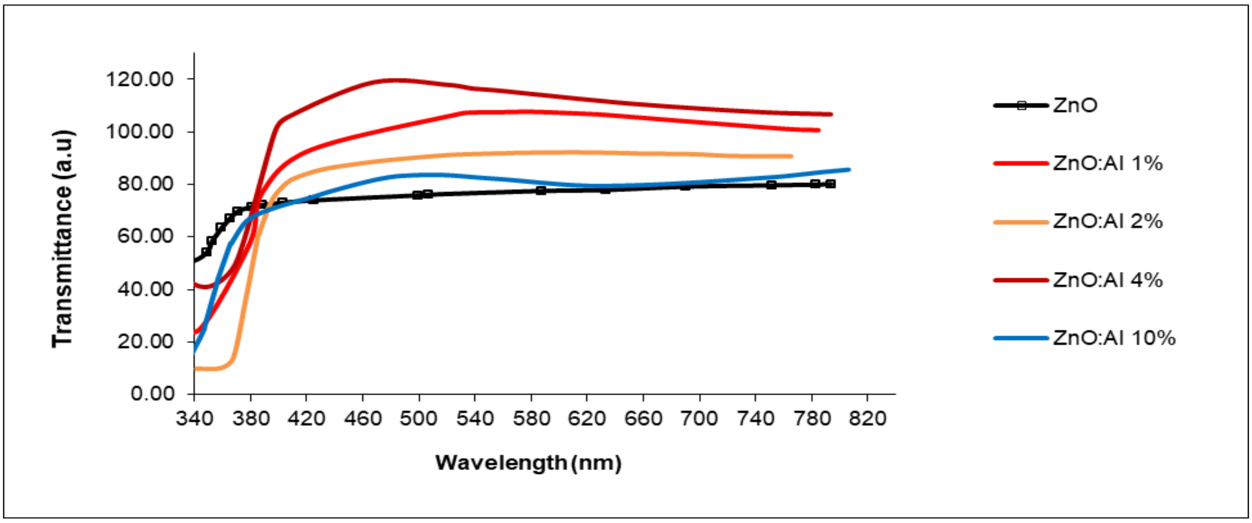

Figure 3 Transmittance behavior of ZnO and Al-doped ZnO thin films.

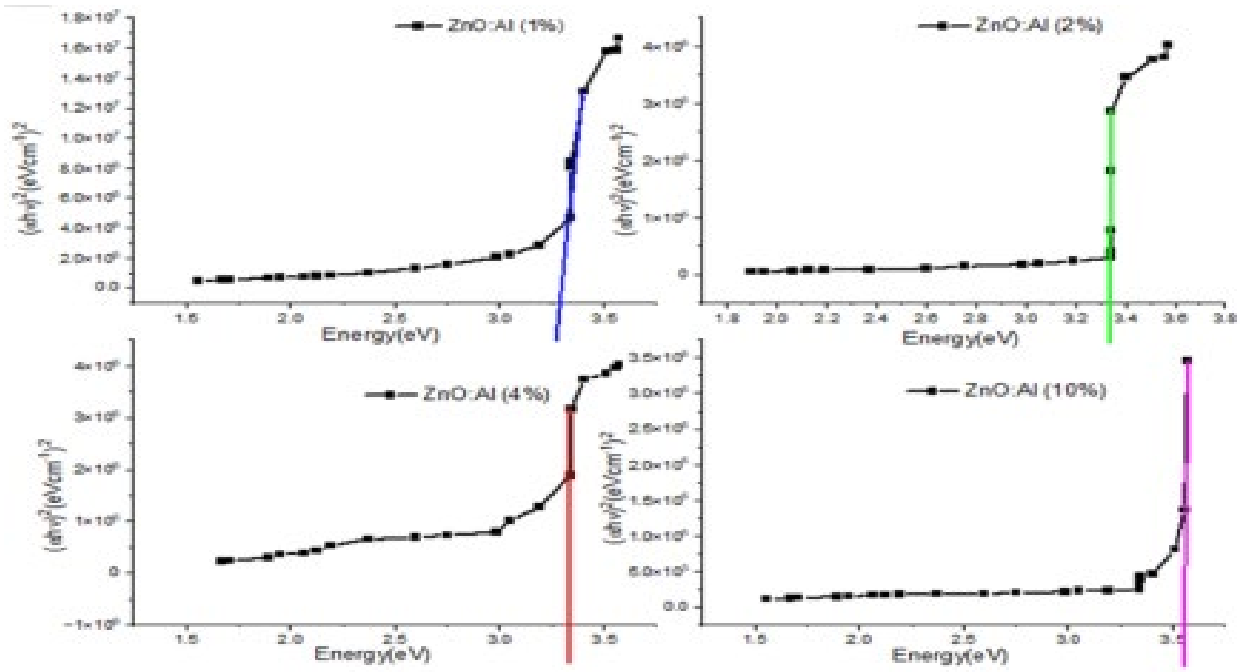

Figure 4 Energy gap of Al-doped ZnO , (1%; 2%; 4%,and 10%) films.

4. CONCLUSIONS

Morphology of (ZnO:Al 4%) films seems to us compact; smoothing surface; and good adhesion. Al/doped ZnO formation hexagonal wurtzite structure; and the absorption of edge of Al/doped ZnO (1%) film's increases with the shift at low wavelength; and becomes less sharp at higher wavelength The ZnO doped with aluminum's absorption edge (2; and 4 %) films is low at low wavelengths. A transmittance of Aluminum doped ZnO (4%) is the only ratio; which showed high light transmission, while optical transmittance of the film's decrease with the increase in Al/doped more than (10%). The energy gup, come to be more than 3eV with higher values of doping ZnO by aluminum particles.

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