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RESEARCH ARTICLE

The effect of thikness on the structural and optical properties of ZnO thin films Prepared by chemical spray pyrolysis methode

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Abstract

ZnO thin films have been prepared on glass substrates at 400°C substrate temperature by the chemical spray pyrolysis(CSP) method using an aqueous solution with varying diffrent thickness. The films have been characterized to evaluate the structure ,transmittance and optical energy band gab. X-ray diffraction(XRD) studies show that the films are polycrystalline with hexagonal structures. The grain size of the films was found increased with increasing the thickness values. The optical band gab energies and types of optical transition and absorption of the films have been determined from the optical transmittance spectra. The optical absorption studies in the wavelength range 400–1100 nm show that band gap energy values of the films decrease from (2.5-2.35)eV as the thickness increases

INTRODUCTION

in ceramic technology and thin film technology due to ZnO powders and thin films are very important materials numerous interesting properties exhibited by this material. ZnO powder with suitable dopants is used as a in sensing combustible a varistor in ceramic technology a sensor element , photoconductor in electrophotography to their valuable Zinc oxide thin films attract much interest due ,[1]and an additive in various ferrites gases high optical transparency in the visible and near-infrared , properties such as chemical stability in hydrogen plasma Due to these properties, ZnO is a promising . the electromagnetic spectrum and high refractive index region of The UV . [2] and transparent conducting material material for solar cell applications such as antireflection coating environmental monitoring, solar astronomy, air quality photodetector has wide range applications such as monitoring, accurate measurement of radiation for the treatment of UV-irradiated skin and missile warning system Therefore, the investigation of dependence of photoresponse properties of sprayed ZnO thin films on precursor . ZnO thin films are grown by different techniques such as pulsed laser deposition (PLD [3]concentration is valuable .[4]magnetron sputtering, MOCVD, spray pyrolysis etc

Experimental

The preparation of membranes (ZnO) using material zinc acetate (Zinc Acetate Dehydrate) and form chemical [(CH3COO) 2Zn .2H2O] and purity (99%) is equipped with a German company, which is about instant white color very small crystalline particles in the water, attended decomposition solution at room temperature and concentration (0.1M) and it dissolves (2.1949g) of material in zinc acetate (100ml) of distilled water, and to get the weight to be dissolved within the past been used titer following relationship :

$$M = (Wt / MWt). (1000 / V) \qquad (1)$$

And mix the solution using a magnetic mixer (Magnetic Stirrer) for min (15-10) to complete the dissolution process, and then leaves the solution for an hour to get a homogeneous solution, and then the solution is placed in the spray device tank and sprayed the solution on the bases heated temperature (400) C ° in the form of payments within a specific time times (for spraying the solution 5 seconds and then stop for 55 seconds), and upon the arrival of the droplets of the solution to the hot-Qaeda surface of the composite steel resulting from the evaporation of water droplets of the solution reacts to the influence of heat to turn into ((ZnO, and constantly continue spraying process chemical reactions to produce membrane (ZnO).

3. Results and Discussion

3.1. X-Ray Diffraction Analysis

The XRD pattern of ZnO thin film fabricated by chemical spray pyrolysis techniqueon glass substrates with thickness (270 , 450 & 690) nm is shown in Figure 1. All the peaks of the ZnO thin films correspond to the peaks of standard ZnO (JCPDS (36-1451). For all the samples, (100), (101) and (002) diffraction peaks are observed in the XRD pattern, showing the growth of ZnO crystallites along different directions. Strong preferential growth is observed along (002) plane indicating that the films are oriented along c-axis [6]. The typical hexagonal wurtzite structure of thin films is inferred from the XRD pattern. it is clear that the ZnO hexagonal structure was dominated for all peaks as matched with the ASTM card of ZnO Similar results have been reported[1],[2],[3],[5],[6],but the Intensity incresing with increse the thickness.

Figure (1-a) XRD pattern of ZnO thin film.

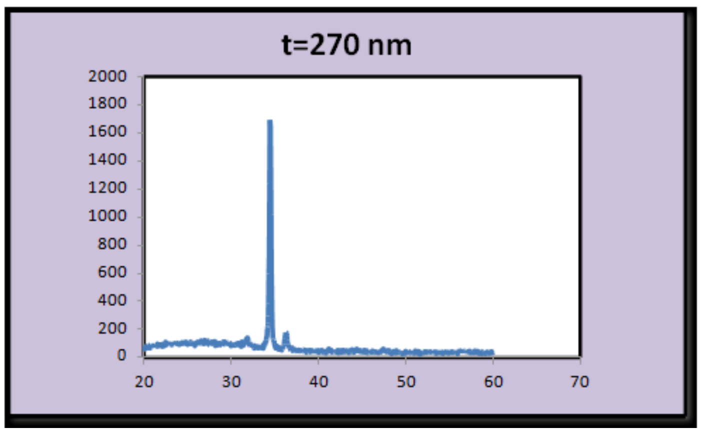


Figure (1-b) XRD pattern of ZnO thin film.

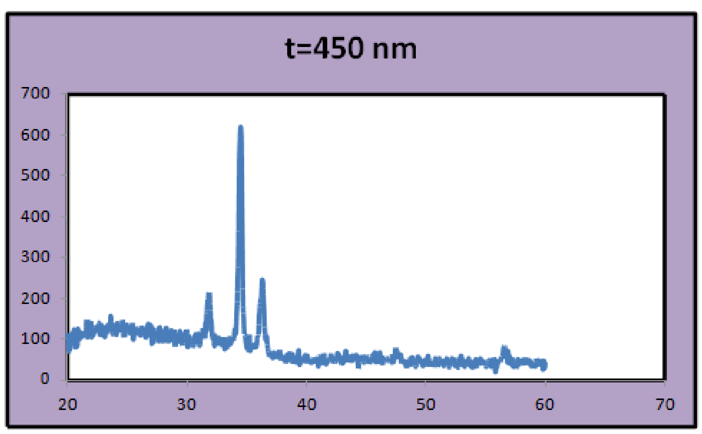
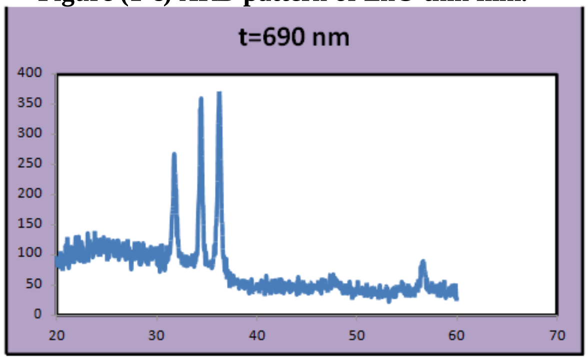


Figure (1-c) XRD pattern of ZnO thin film.



The lattice constant ,a and c, are evaluated from the following relations[7]

$1/d = [(4/3)\{(h^2+hk+k^2)a^2]+(l^2/c^2)$ ……………….(2)

The calculated values of lattice constants for SnO2 thin films are in good agreement with ASTM data . Similar results have been reported by [1],[15].

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It is clear from XRD patterns of for ZnO thin films were 2.5 eV , 2.45 eV , 2.35 eV respectively. thin films , that the full width at half maximum (FWHM) decreases with the increasing of thikness in these films. This decreasing in (FWHM) indicates an increasing in the grain size of ZnO thin films .T**h**e crystallites sizes (D) of the films are estimated using the Scherer formula [8]:

where k is a constant taken to be 0.9, λ is the wavelength of X-Ray used (λ = 1.54 Å) and β2θ is the full width at half maximum of (002) peak of XRD pattern, Bragg angle, 2θ, is around 34.44°. The average value of grain size is found to be 20 nm. The dislocation density (δ), defined as the length of dislocation lines per unit volume, are estimated using the equation[9]:

δ=

The values of the dislocation density of SnO2 thin films are given in table (2) . It is evident from this table that the dislocation density of for ZnO thin films were 2.5 eV , 2.45 eV , 2.35 eV respectively. thin films decreases with increasing in thikness which can be also deduced from the increasing in the grain size where the dislocation density is proportion reversely with the square of the grain size according to eq.(3). This decreasing in the dislocation density indicates an improvement in the crystallinity of for ZnO thin films were 2.5 eV , 2.45 eV , 2.35 eV respectively. thin films and their homogeneity as increasing in thikness.

The strain (ε) developed in for ZnO thin films were 2.5 eV , 2.45 eV , 2.35 eV respectively. thin films can calculated from the relation [10] :

ε

The origin of strain is related to lattice misfit which in turn depends upon the growing condition of the films. The values of the strain of for ZnO thin films were 2.5 eV , 2.45 eV , 2.35 eV respectively.thin films are given in table (2). It is clear from this table that the strain in SnO2thin films decreases with increasing in thikness which can be also deduced from the increasing in the grain size, where the decreasing in
the strain and the dislocation density with the increasing in
the grain size is a well-known phenomenon. In addition the films with lower strain and dislocation density improves the stoichiometry of the films which inturn causes the volumetric expansion of the films.

Growth of pure

Table (1) THE STRUCURAL PARAMETERS OF ZnO thin film

X	(2θ Exp .(deg	2θ Stan (.(deg	(d Exp.(A°	(d Stan.(A°	(hkl)
270	34.138 34.488 36.289	31.769 34.421 36.252	2.624 2.598 2.473	2.8143 2.6033 2.4759	100 002 101
450	31.8316 34.4885 34.4885	31.769 34.421 36.252	2.809058 2.598502 2.474382	2.8143 2.6033 2.4759	100 002 101
690	31.7853 34.4173 36.2551	31.769 34.421 36.252	2.813059 2.603684 2.475814	2.8143 2.6033 2.4759	100 002 101

nO thin films prepared by chemical spray

pyrolysis on silicon TABLE (2)VARIATION OF THE FULL WIDTH AT HALF MAXIMUM , GRAIN SIZE , LATTICE CONSTANTS, DISLOCATION DENSITY AND STRAIN OF ZnO THIN FILMS WITH TIKNESS

thikness	(FWHM) rad	Grain size (nm)	Lattice Constants		δ(lines.m^{-2}) x 10^{16}	ε (rad) x 10^{-4}
			a(Å))	c(Å)		
270	0.025382	056	3.2481275	5.19700	0.1844962	3
450	0.020951	4	3.2481275	5.19700	0.1844962	26
690	0.015176		3.254605	5.20736	0.0145584	25

,[2] ar element in sens

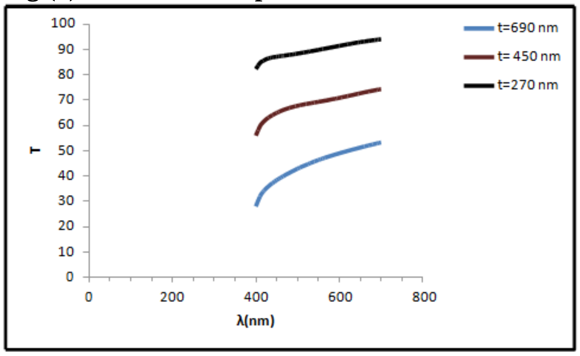
i 3.2.Optical properties

The transmittance spectrum of ZnO thin films where thikness equal (270 , 450 & 690)nm are shown in Fig.(2). All spectrums reveal very pronounced interference effects for photon energies below the fundamental absorption edge by exhibiting interference pattern . Such behavior of the spectrum is evidence of the thickness uniformity of the films.It is clear from the same figure that the transmission decreases with increasing in thikness. In addition the variation of the transmittance of ZnO thin films with the wavelength is very important because this variation will limit the transmitted wavelengths which play an important role in determination the category/type of the optical filters . The absorbance spectrums of ZnO thin films where x equal (270 , 450 & 690)nm are shown in Fig. (3). It is clear that as the thikness increases the absorbance of ZnO thin films is increased . which results in an increase of the depth of donor levels associated with these vacancies and these levels will be available for the photons to be absorbed therefore the absorbance of ZnO thin films will increase with increasing in thikness . As well as from the same figure , it can be seen that the absorption edge shifts to the higher wavelengths corresponding to the red region as the Se concentration increased .

ng combustible gases [3] and

an

Fig.(2) Transmission spectrum of ZnO thin film.



thin films ZnO Fig.(3) Absorbance spectrums of

Figure (4) illustrates the absorption coefficient of ZnO thin films at (270 , 450 (690 & nm. It is clear from this figure that the absorption coefficient of ZnO thin films increase with increasing in thikness .This increase is attributed to the increase in the absorbance of ZnO thin films with the increase with increasing in thikness where the relation between the absorbance and the absorption coefficient is proportional at constant thickness according to eq.(6[20])((6)

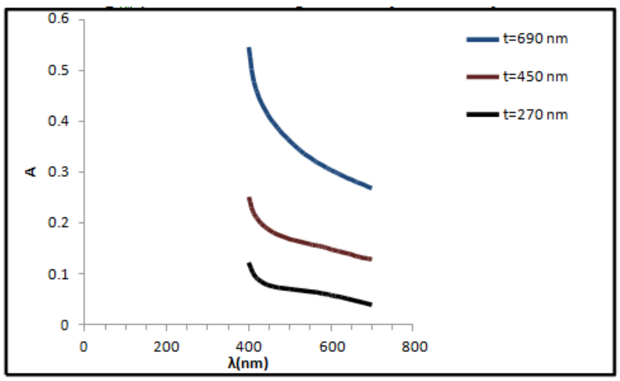


Fig.(3) Absorbance spectrums of ZnO thin films

thin films ZnO of absorption coefficient *(4).*Fig

have method spray pyrolysis by chemical for ZnO thin films prepared (The optical energy gap values (E_g by using of these films been determined from the region of the high absorption at the fundamental absorption edge :[19] Tauc equation

(7) ....

Where , is the absorption coefficient , is the incident photon energy in eV , is a constant depends on the nature of the material (properties of its valence and conduction band) [19] and is a constant depends on the nature of the transition between the top of the valence band and bottom of the conduction band.

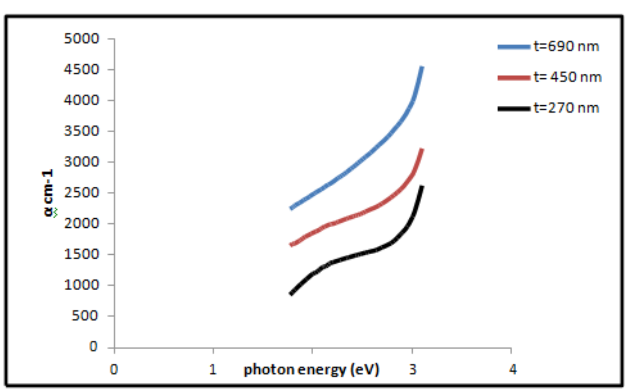


Fig.(4) absorption coefficient of ZnO thin films

This equation is used to find the type of the optical transition by plotting the relations $(\alpha h\upsilon)^2$, $(\alpha h\upsilon)^{2/3}$, $(\alpha h\upsilon)^{1/2}$ and $(\alpha h\upsilon)^{1/3}$ versus photon energy (hυ) and select the optimum linear part. It is found that the first relation yields linear dependence, which describes the allowed direct transition , then E_g was determined by the extrapolation of the portion at (α=0) as shown in Fig.(7). It is clear that the optical energy gap for ZnO thin films decreases as the thikness of the films increased. The optical energy gap values for ZnO thin films were 2.5 eV , 2.45 eV , 2.35 eV respectively.

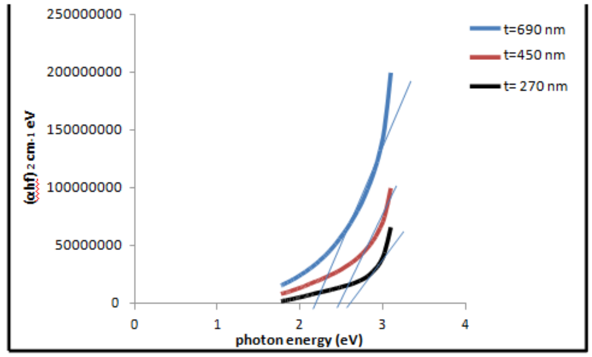


Fig.(5) (· hυ)2 as a function of hυ for ZnO thin films
(a) t=270 , (b) t=450 , (c) t=690

Fig*) (5).*hÅ) *2*as a function of h*υ* for ZnO thin films

(a) t=270 , (b) t=450 , (c) t=690

:[have been determined by using the following equation [19 () The extinction coefficient

......................................(8)

. is the absorption coefficient andis the wavelength of the incident photon : Where illustrates the variation (6) .giF . α ot roivaheb ralimis a sah dna αdepends on It is clear from this equation that nm . From this (690 & 450 , 270) for thikness photon energy thin films with ZnO of the extinction coefficient of varies slightly with the increasing in the photon energy corresponding to the decreasing figure , it can be noted that increases highly at the absorption edge region and this increasing is attributed to in the absorption coefficient. Then reaches to its maximum the increasing of the absorption coefficient due to the direct electronic transitions thereafter value at the high absorption region corresponding to the increment in the photons' energy and the increasing in the absorption coefficient with the decreasing in the wavelength . In addition , it is clear from this figure that with the increasing in the absorption increases . This is attributed to the increasing in the thikness the extinction coefficient coefficient due to the increasing of the depth of donor levels associated with tin vacancies and these levels will be available for the photons to be absorbed causing an will increase with the increment in the absorbance and leads to increase in the absorption coefficient. Therefore . ti no sdneped dna αincreasing in the thikness since it has a similar behavior to

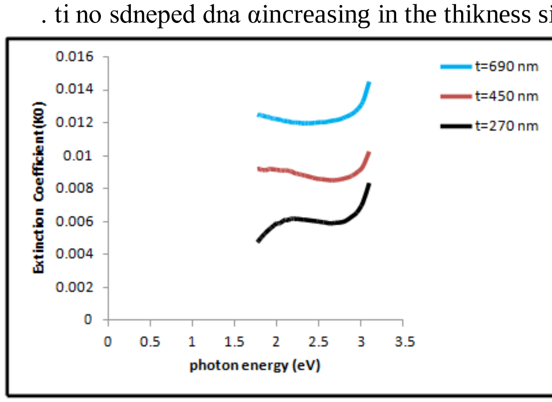


Fig.(6) Extinction coefficient as a function of photon energy
for ZnO thin films

Fig *(6).*Extinction coefficient as a function of photon energy

for ZnO thin films

:[thin films have been determined by using the following equation [19 ZnO The refractive indexof

................(9)

.extinction coefficient the is : Where , R : is the reflectance of the films and k_o

It .(7) .is illustrated in Fig thin films ZnO The variation of the refractive index as a function of the photon energy for Also it can be .photon energy in the increases is clear from this figure that the refractive index increasing with the with the increasing in the thikness . This increasing is increases thin films ZnO observed , that the refractive index of to the increasing in the grain size of the films with the increasing in the thikness which interns causing an attributed increment in the compactness of the films which in turns reduces the speed of light in the material of the thin film varies according to the grain size even if the () and then leads to an increasing in the refractive index. Where crystalline structure is itself of the material

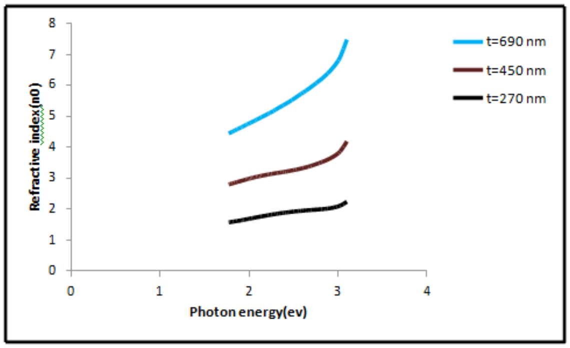


Fig.(7) The variation of the refractive index as a function of the photon energy for ZnO *thin films*

Fig *(7).*The variation of the refractive index as a function of the photon energy for ZnO thin films

figure(8)It is clear that the real part of the dielectric constant of ZnO thin films increase with the thikness increases due to the dependence of the real part of the dielectric constant on the refractive index values , where the refractive index increase with the thikness increases.

figure (9) . It is clear that the increase in the thikness leads to an increase in the imaginary part of the dielectric constant of ZnO thin films . This is attributed to the dependence of the imaginary part of the dielectric constant on the extinction coefficient values , where the extinction coefficient increases with the increase in the thikness.

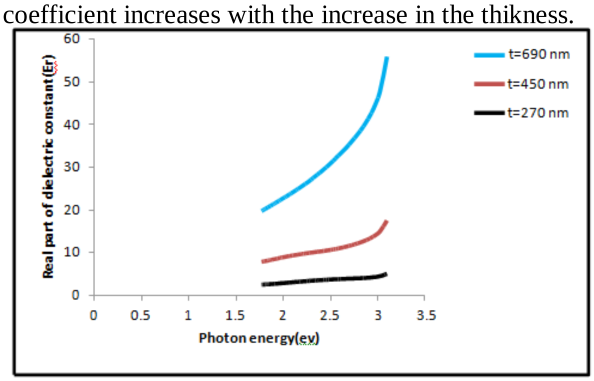


Fig.(8):The real part of the dielectric constant of ZnO *thin films*

thin films ZnO The real part of the dielectric constant of*:(8).*Fig

Fig*:(9).*The imaginary part of the dielectric constant of ZnO thin fil m

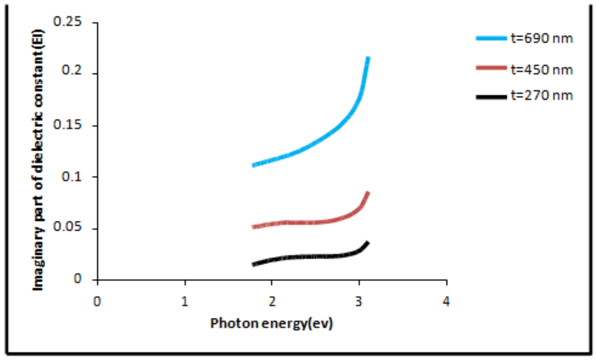


Fig.(9):The imaginary part of the dielectric constant of ZnO *thin film*

.3.3Conclusion

In this study, ZnO films are deposited by spray pyrolysis technique. The structural, optical properties of ZnO thin films were investigated as a function of precursor concentration. The films are polycrystalline with hexagonal structure with (002) peak as a preferred orientation. The grain size of ZnO films increases with increasing in thickness. The optical transmittance spectrum shows that transmission decreas with increasesing the thickness and the optical energy gap for ZnO thin films decreases as the thikness of the films increased. The optical energy gap values for ZnO thin films were 2.5 eV , 2.45 eV , 2.35 eV respectively.

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