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Research Article

OPTICAL PROPERTIES OF SOL GEL DIP COATED SnO_2 FILMS

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ABSTRACT

In this paper we have reported on dip coating SnO_2 films prepared on clean glass substrate post heat treated at different temperature in the range of 350-525°C. X-ray diffraction pattern indicate the films were single phase tetragonal structure. The room temperature PL spectra of the samples show that an intensive Ultra Violet peak at 390 nm (about 3.18eV) and a shoulder at 430 nm (about 2.88eV) are observed in two different temperatures (450°C and 525°C). In addition there is a broad peak at 520 nm (about 2.38eV). Transmission spectra exhibit good transparency in the visible region. The films thickness was estimated using surface profilometer and found to be in the range of 500-640 nm.

The value of some important parameters refractive index (n), extinction coefficient (K) and dielectric constant (ε) of SnO_2 thin films are determined from the transmission spectra. The energy band gaps of the films are found to vary from 3.68 eV to 3.88 eV. Refractive index was in the range of 2.05-2.18. The dispersion of refractive index was analyzed by W.D single oscillator method. The optical band gap (Eg), dispersion energy (E_d) single oscillator energy (E_o) and optical conductivity were estimated and these films would be suitable for the application of solar cells and sensors.

INTRODUCTION

Transparent conductive oxide (TCO) thin films have figured prominently for many years in a wide variety of application such as gas sensor, biosensor and transparent electrode heating elements in aircraft windows for deicing and defogging, antistatic coating on instrument panels, electrical contacts in liquid crystal, electro chromic and electroluminescent display. The material has high reflection in the infrared region in conjunction with high transparency in the visible region. [1-2] The revealed research interest in these films in recent years stems from the possibility of fabricating large area, stable and high efficiency solar cells utilizing these films. SnO_2 is a promising material due to wide band gap (3.6 - 4 eV) and good electrical conductivity. More over among them SnO_2 thin films is one of the most significant TCO materials. It is used as transparent electrode, transparent windows in liquid crystal display, gas sensor, solar cells [3-5]. There are several techniques to prepare SnO_2 films such as electrostatic spray deposition [6] pulsed laser deposition[7-8] laser chemical vapor deposition[9] and sol gel[10-11]. Among these techniques sol gel dip coating provide an easy route for the preparation of the thin films at a low cost. The structural morphological compositional and optical properties of the prepared thin films were studies using Phillips X-ray unit and cukα radiation and U3400 Hitachi UV–Vis-NIR spectrometer. Photoluminescence studies were made at room temperature using a Hitachi X-ray fluorescence spectrometer. Transport parameters were determined by Hall measurement. The resistivity of the films was estimated by the four probe method. Single oscillator model is used to find the determination of optical parameters.

Experimental studies

Tin Oxide thin film preparation was given in our earlier paper [12]. After the preparation films it was characterized by different studies and their results was reported.

RESULT AND DISCUSSION

Fig (1) shows XRD pattern of the SnO_2 thin films formed at different temperature in the range of 350-500°C. All the peaks are corresponding to single phase tetragonal structural. The grain size increases from 10 nm to 25 nm with increase of formation temperature. XRD pattern revealed that the deposited films were found polycrystalline in nature with tetragonal structure, and also the microstructure parameters such as crystallite size, strain, and dislocation density and microstrain value reported in the previous paper [13]. Fig. 2 shows the transmittance spectra of the SnO_2 thin films on

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glass substrate in the wavelength range of 300-800 nm. The films exhibited good transparency with interference fringes owing to the phenomenon of multiple reflections SnO_2 films are n-type materials their absorption properly is a very important optical parameter. Optical absorption measurement is used to find the optical parameter. The optical data is used to plot a graph of hv versus $(\alpha hv)^{1/2}$. Extrapolation of the plots to the x-axis gives the band gap energy of the SnO_2 film deposited at various temperatures. The amplitude of atomic vibrations increase with temperature resulting in larger atomic spacing. The band gap is affected by a smaller extent due to the interaction between the free electrons, holes and lattice phonons. One of the mechanisms for electrons to be excited to the CB is due thermal energy and the conductivity of semiconductor is strongly dependent on the temperature of the materials. The band gap energy of SnO_2 thin film deposited at optimized deposition condition is 3.68 eV and this value as in good agreement with the value reported earlier. [14]

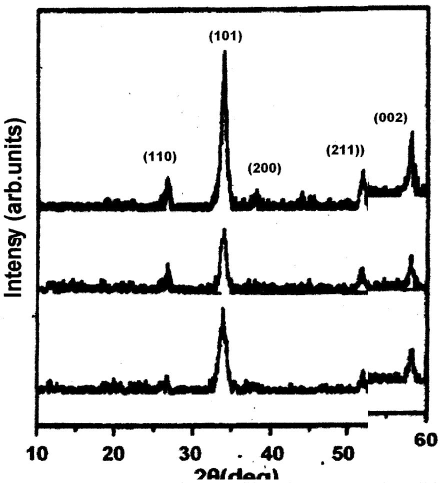

Figure 1 - XRD patterns of SnO_2 films heat treated at different temperatures (a) 350˚C (b) 450˚C (c) 525˚C

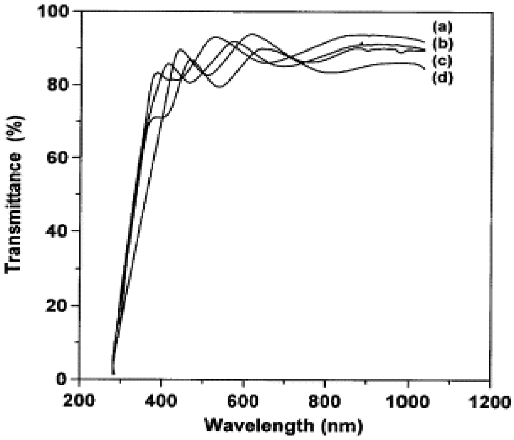

Figure 2 Transmission spectra of SnO_2 films heat treated at different temperatures (a) 350˚C (b) 450˚C (c) 525˚C

$\alpha hv = A(h\upsilon - E_g)^{-1/2}$-----------------------------(1)

where α, A is the absorption constant, constant of the equation and hv is the incident photon energy. The experimental observation of transmission spectra was used to evaluate the refractive index and extinction coefficient using the procedure described in reference [15]

$N = [N + (N^2 - s^2)^{½}]^{1/2}$ ----------------(2)

$N = 2S\,(T_{max} - T_{min}) / (T_{max}.T_{min}) + (s^2+1)/2$- (3)

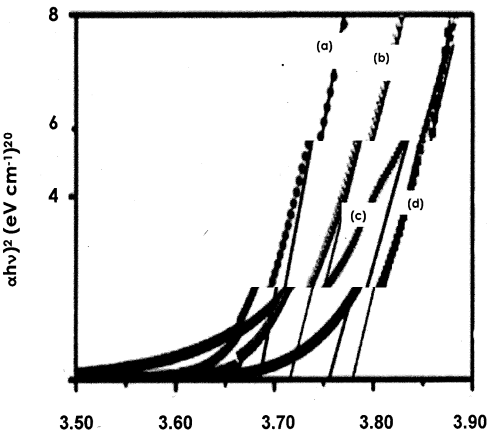

Figure 3 shows $(\alpha hv)^2$ vs. hv plot of SnO_2 films formed at (a) 350˚C (b) 450˚C. (c) 500˚C (d) 525˚C

S is the refractive index of the substrate (glass-1.5) T_{max} and T_{min} are the maxima and minima transmittance at the same. The maximum value of refractive index is observed at 2.18. The refractive index variation may be due to the films thickness as shown in fig.4.Fig.5 shows the room temperature resistivity, mobility and carrier concentration in SnO_2 films as a function of formation temperature respectively. The resistivity decreases with increase of temperature and attains a constant value. This decrease is due to an increase carrier concentration. The mobility decrease with formation temperature. The resistivity values are higher than chemical vapor deposition tin oxide films by Kajima.*et al* [16] the spray pyrolysis techniques by patil.*et al* [17 and the commercial film by Napoo *et al* [18]. Our Hall mobility values are lower than the value measured in single crystalline samples. Grain boundaries in the films limit the carrier transport which is responsible for the high resistivity values observed in this study.

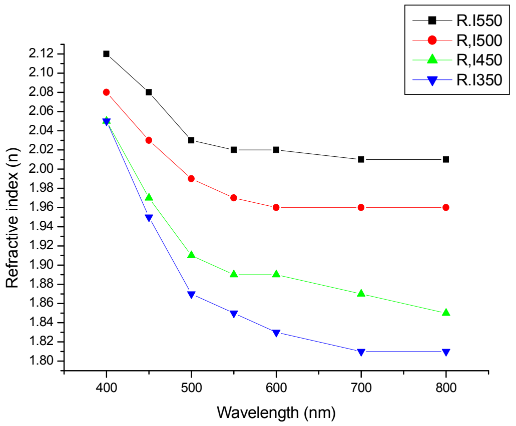

Figure 4 shows the variation of refractive index with different formation temperature

Room temperature PL spectra produce an intensive U-Violet peak at 390 nm (about 3.18eV) and a shoulder at 430 nm (about 2.88eV) are observed in fig.6. In addition there is a broad peak at 520 nm (about 2.38eV). Oxygen vacancies are the common defects observed in polycrystalline oxides which are responsible for the formation of donor levels [19] Existence of oxygen vacancies in our films are responsible for the resistivity values observed in this study. As a result of in-diffused oxygen recombining with oxygen vacancies due to annealing under ambient conditions decreased [20-21] from fig.8. It can be seen that the Intensity of the UV peak increases obviously as the heat treatment temperature increases and the decrease of the number of oxygen vacancies does not lead to the decrease of the Intensity of the ultra violet peak, which shows that the oxygen vacancies donor play an important role in the origin of the Ultra-violet, Extinction coefficient (k) of SnO_2 films are estimated by the following expression

$K=\alpha \lambda / 4 \times \pi$ -------------- (4)

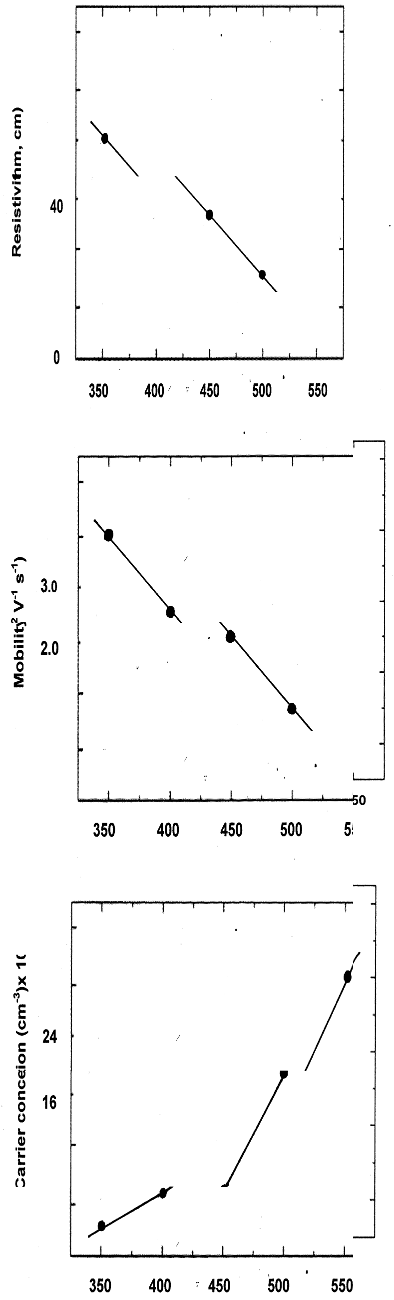

Figure 5 shows the variation of Resistivity, mobility, carrier concentration with formation temperature (Room Temp.)

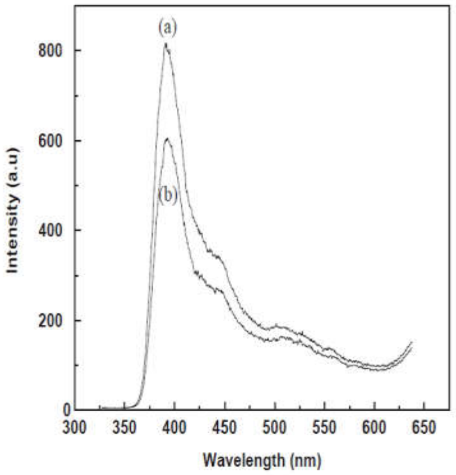

Figure 6 Room temperature photoluminescence spectra of SnO_2 films formed at different temperatures (a) 525˚C (b) 450˚C

Fig 7-8 shows the variation of complex dielectric coefficient of sol gel dip coated SnO_2 thin films deposited on glass substrate. As solar cell materials, dielectric films are used to form electrical and Interfacial barriers between metal and semiconductor, for passivation and for antireflection coatings. The real and imaginary parts of the dielectric constant were determined using the relation

$\varepsilon = \varepsilon_r + \varepsilon_{i} = (n + ik)^2$ ---------------------- (5)

where ε_r and ε_i are the real and imaginary parts of the dielectric constant respectively.

$\varepsilon_r = (n^2 - k^2)$-----------------------------(6)

and $\varepsilon_i = 2nk$ ----------------------------------(7)

The fundamental intrinsic property of a material is the complex dielectric constant. The property responsible for slowing down the speed of light is the real part of the dielectric constant. The high refractive index value observed in this study is the reason for the maximum value of the real part of the dielectric constant. While, dispersion is related to the real part of dielectric constant, dissipative rate of the wave is provided by the imaginary part.

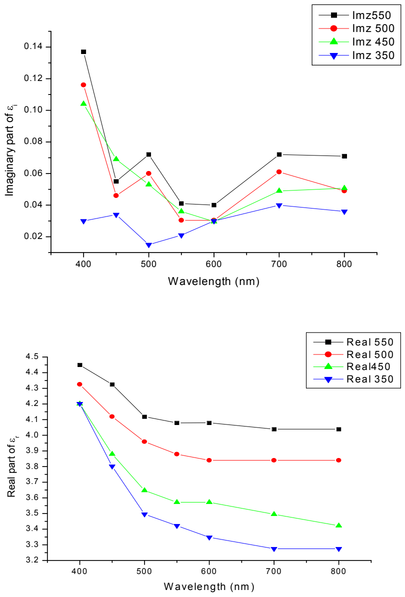

Figure 7 & 8 Real and Imaginary part of dielectric constant of SnO_2 thin films with different temperature

The optical conductivity is determined (table-1) using the usual relation

$$C = \alpha\ n\ c \text{ ------------------ } (8)$$

Table 1 The optical parameter of SnO_2 thin films

Real part of ε_r	IMZ part of ε_i	Optical conductivity
≈4.167	≈0-069	≈5.12 x 10 ^{13}

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Fig.9 display the variation of optical conductivity of tin oxide thin films deposited on glass substrate, in the case SnO_2 thin films optical conductivity value is observed at 5.12 x10 ^{13} for glass substrate. The high absorbance of the tin oxide films at high photon energies is the reason for the high optical conductivity. The dispersion phenomenon is an important factor in designing optical communication devices for spectral dispersion.

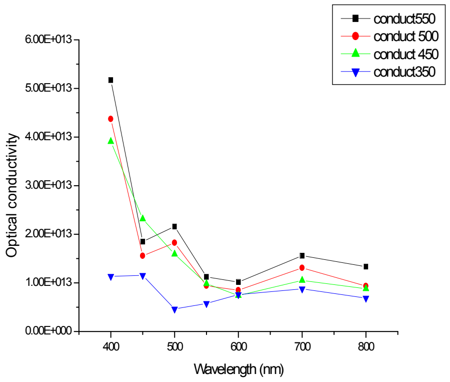

Figure 9 shows the variation optical conductivity with different temperature.

Although these rules are quite different in detail, one common feature is the over whelming evidence that both crystal structure and iconicity influence the refractive index behavior of solids in ways that can be simply described. The single oscillator model [22] can be used to evaluate the spectral dependence of the refractive index of many semiconductors. The dispersion data of the refractive index was evaluated using the above single effective oscillator model [22–23]. In the low absorption region, the refractive index using single oscillator model is given by

$$h^2 (hv) = 1 + E_d E_o / (E_o{}^2 - (hv)^2) \text{ ----------------}(9)$$

The average energy gap is represented by E_0. The band gap energy E_g and E_o are related through the approximation obtained from the experimental results as $E_o = 2\ E_g$. From a plot of $(n^2 - 1)^{-1}$ and $(hv)^2$, the values of E_o and E_d can be estimated from the slope and intercept of the plot.

Table 2 The optical parameter of SnO_2 thin films

Sample	E_g (eV)	E_o (eV)	E_d (eV)
At 350˚C	3.68	7.36	14.65
At 450˚C	3.72	7.44	14.81
At 500˚C	3.75	7.50	14.93
At 550˚C	3.78	7.56	15.05

CONCLUSION

SnO_2 thin films were synthesized by sol gel route on glass substrate. The effect of thickness of tin oxide thin film, structural, electrical and optical properties has been studied. It was clear that the structural optical and electrical properties strongly depend on the films thickness.

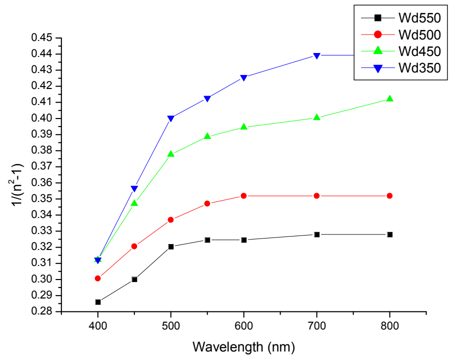

Figure 10 shows $(n^2\text{-}1)^{-1}$ versus (λ) curve.

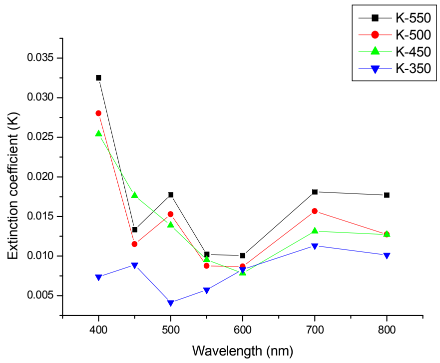

Figure 11 Variation of extinction coefficient (k) as a function of Wavelength and the refractive index as a function of wavelength..

With increase in thickness of the films, the transmittance and resistivity decrease. An increase in refractive index is observed with thickness. The increase in the band gap energy results in an increase of the dielectric properties of the film**.** The real and imaginary parts of the dielectric constant increase with increase in photon energy. The optical conductivity also increases with increase in photon energy and this can be attributed to the increase in absorption coefficient with increase in photon energy. Finally an attempt was made to apply W.D. single oscillator model to calculate the optical parameters. The optical band gap obtained by Tauc's extrapolation and the static refractive index from Cauchy fitting were is good agreement with W.D.model.

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