

# Properties of pure and light antimony-doped tin oxide thin films prepared by e-beam technique

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Transparent conducting antimony-doped tin oxide SnO<sub>2</sub>:Sb films were prepared on glass substrates by e-beam evaporation technique. The effect of Sb-content and annealing temperature on the optical, electrical and structural properties of the formed films were investigated. The as-deposited films showed amorphous structure and low transmittance that increased with increasing Sb-content. Upon annealing, the film ordering enhanced. The annealed SnO<sub>2</sub><sub>100-x</sub>Sb<sub>x</sub> (x= 3 wt%) films showed the minimum resistivity of  $\sim 6 \times 10^{-3} \Omega \text{ cm}$  at temperature 400 °C and its transmittance exceeded 84 % in the visible region. The other parameters such as free carrier concentrations, refractive index, Urbach energy were calculated for as-deposited and annealed films.

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## 1. Introduction

Undoped tin oxide (SnO<sub>2</sub>) is a wide band-gap semiconductors material with  $E_g \sim 3.6 - 4 \text{ eV}$  [1-4]. SnO<sub>2</sub> in the form of thin film is a transparent material, characterized by high optical transmission in the visible region [5] and low resistivity. Particularly, they are stable up to high temperatures, have excellent resistance to strong acids and bases at room temperature as well as to mechanical wear and they have very good adhesion to many substrates[6]. Therefore, this material is used as transparent electrodes in many applications, such as gas sensors, solar cells, liquid crystal displays, etc [7-11]. In addition, SnO<sub>2</sub> films are more chemically stable than other TCOs such as zinc oxide (ZnO) and Sn-doped In<sub>2</sub>O<sub>3</sub> (ITO) [12].

SnO<sub>2</sub> thin films can be prepared using various techniques, such as pulsed laser deposition [13], chemical vapor deposition [14], sol-gel [15] and radio frequency magnetron sputtering [16].

It was found that the electrical resistivity of these films decreases by doping with F<sup>-</sup> or Sb<sup>5+</sup> without significant changes in their optical transmittance [17-19]. Beside, it was reported that the small amount of Sb dopant results a good electrical conductivity of SnO<sub>2</sub> films without losing optical transparency. However, large additions diminish the conductivity, optical transparency and crystallinity of these films [20, 21].

In this paper, the effect of small amount of Sb dopant and heat treatment on the structural, optical and electrical properties of SnO<sub>2</sub> thin films deposited by electron beam evaporation technique were investigated.

## 2. Experimental details

For preparing SnO<sub>2</sub><sub>100-x</sub>Sb<sub>x</sub> (x = 0, 1, 3, 5, and 8 wt%) bulk samples, a highly pure (99.999% purity) powder of SnO<sub>2</sub> and Sb were ground separately and sieved. A mixture of SnO<sub>2</sub><sub>100-x</sub>Sb<sub>x</sub> with different x ratio was prepared. To increase complete mixing, the mixtures were ground for at least two hours. Then, they were made into tablet form using a cold pressing technique. In order to increase the diffusion process and consequently improve the homogeneity of the material, the sintering was performed for the present mixtures at 900 °C for 4 h.

Thin films of the considered ratios were prepared by electron beam evaporation in an Edward's high vacuum coating unit model 306 A under pressures of  $5 \times 10^{-6}$  Torr. The films were prepared on unheated ultrasonically cleaned microscopic glasses. The thickness of the films ( $\approx 100 \text{ nm}$ ) was controlled using a digital film thickness monitor model TM 200 Maxtek. The deposition rate was  $\approx 10 \text{ nm/min}$ . The post-thermal annealing of the as-deposited films was carried out in a fully controlled furnace in air for 30 min. The annealing temperature range was from room temperature to 550 °C.

The crystallinity of the as-deposited and annealed films was examined using a Philips-PW1710 X-ray diffractometer. CuK $\alpha$  radiation ( $\lambda = 1.541838 \text{ \AA}$ ) was used from the X-ray tube with normal focus. The optical transmittance ( $T$ ) and reflectivity ( $R$ ) of SnO<sub>2</sub><sub>100-x</sub>Sb<sub>x</sub> films were measured using a Jasco 570 double-beam spectrophotometer over a wavelength( $\lambda$ )range of 200–2500 nm at normal incidence.

The resistivity measurements were carried out using a two-terminal configuration by applying a constant voltage ( $\approx 5 \text{ V}$ ) to the sample and measuring the current through it using a Keithley 614 electrometer. The measurements

were done at room temperature. Electrical contacts were made by applying silver paste over the surface of the films with a separation of 3 mm.

### 3. Results and discussion

#### 3.1. Structural properties

The crystalline structure of un-doped SnO<sub>2</sub> and SnO<sub>2</sub>:Sb films is shown in Fig. 1. The X-ray diffraction patterns of as-deposited films are plotted in Fig.1-a. All the patterns show the amorphous structure with some peaks (020), (111), (220) of SnO. Moreover at low Sb-ratio, the peaks of (111) and (042) of Sb<sub>2</sub>O<sub>4</sub> and Sb<sub>2</sub>O<sub>3</sub>, respectively can be observed. Fig. 1-b represents the X-ray diffraction of SnO<sub>2</sub><sub>100-x</sub>Sb<sub>x</sub> (x =, 3 wt%) films annealed at 300 and 450 °C. From this figures it can be observed that, the annealed film at 300 °C shows the peaks (042) of Sb<sub>2</sub>O<sub>3</sub> and (111), (220)of SnO. With increasing the annealing temperature up to 450 °C, the films show an amorphous matrix containing small randomly oriented crystals. Since the peaks (110), (101), (211) of SnO<sub>2</sub> are observed with intensities higher than those of SnO, Sb<sub>2</sub>O<sub>3</sub> and SbSn peaks.

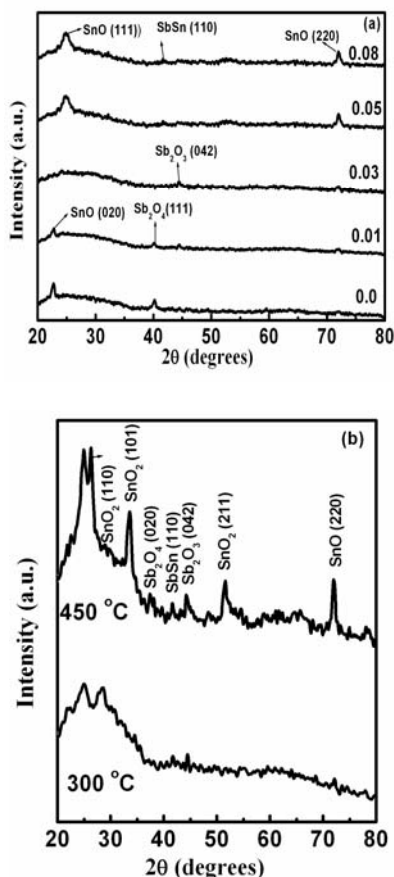


Fig. 1. X-ray diffraction of a) as-deposited SnO<sub>2</sub><sub>100-x</sub>Sb<sub>x</sub> (x =, 0, 1, 3, 5, 8 wt%) films and b) annealed SnO<sub>2</sub><sub>100-x</sub>Sb<sub>x</sub> (x =, 3 wt%) films at 300 and 450 °C.

#### 3.2. Optical properties of as-deposited films

The optical transmittance for as-deposited SnO<sub>2</sub>:Sb films in wavelength range 200-2500 nm is shown in Fig.2. From the figure, it is seen that, the transmittance increases with increasing the Sb-content up to x=5 and then starts to decrease with further increase in Sb-content. The maximum transmittance value is approximately 73 % in the visible region of x=0.05. The transmittance of pure SnO<sub>2</sub> films is significantly low this may be due to the lack of oxygen resulting during the electron beam evaporation process. This result agrees with that in ref [22].

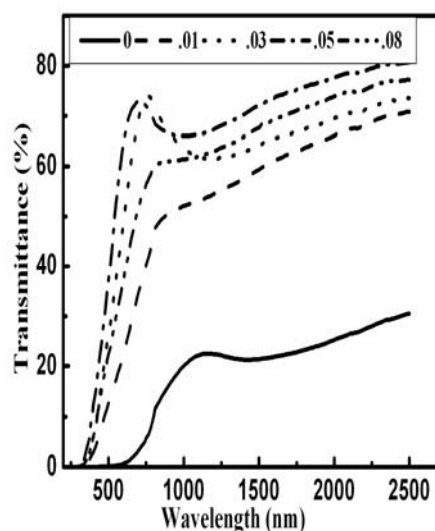


Fig. 2. Transmittance spectra ( $T\%$ ) of as-deposited SnO<sub>2</sub><sub>100-x</sub>Sb<sub>x</sub> (x = 0, 1, 3, 5, and 8 wt%) films.

In order to estimate the optical band gap energy of as-deposited films, the absorption coefficient ( $\alpha$ ) were calculated using the measured transmittance and reflectance spectra [23]. Then, the optical energy gap ( $E_g$ ) was obtained by extrapolating the linear region of  $ah\gamma$  according to the following relation [24]:

$$\alpha h \gamma = A (h \gamma - E_g)^n \quad (1)$$

where  $A$  is a constant,  $h\gamma$  is the photon energy and  $n$  is the factor depend on the nature of electronic transition. The films prepared in the present study have direct allowed transition ( $n = 1/2$ ) as shown in Fig.3-a. The values of optical energy gap was plotted as a function of Sb-content as shown in Fig.3-b. It is observable that, in general,  $E_g$  increases with increasing the Sb-content. The increase in the energy gap can be correlated with the Burstein shift. Since the absorption edge of the films shifts to shorter wavelength as shown in Fig. 2. A similar result was noted by X. Feng et al, when they prepared SnO<sub>2</sub>:Sb films on  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> (0001) substrates by MOCVD [25]. Besides, the width of localized states can be determined from Urbach energy ( $E_U$ ) according to the following equation [26]:

$$\alpha = \alpha_0 \exp\left(\frac{h\nu}{E_U}\right) \quad (2)$$

where  $\alpha_0$  is a constant and  $E_U$  is the Urbach energy. The values of  $E_U$  were calculated and plotted in Fig.3-b. From this figure, it is clear that, the Urbach energy has an opposite behaviour of energy gap. This may be due to the decrease in the film disordering with increasing Sb-content as shown in Fig.1-a.

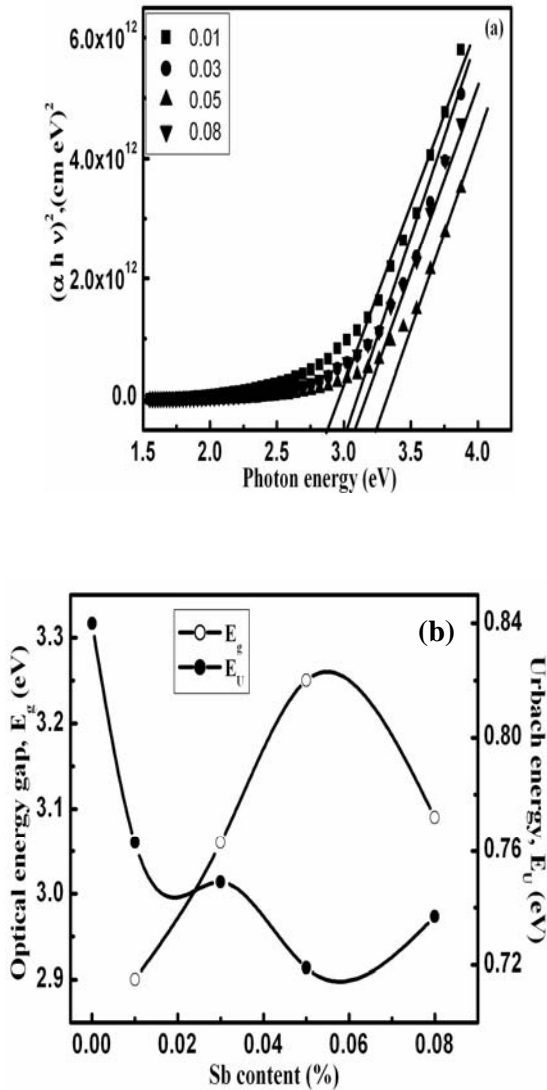


Fig. 3. a)  $(\alpha h\nu)^2$  as a function of photon energy and b) the calculated optical energy gap and Urbach energy of as-deposited  $\text{SnO}_2_{100-x}\text{Sb}_x$  ( $x = 0, 1, 3, 5,$  and  $8$  wt%) films.

The optical free carrier concentration ( $N$ ) was calculated from the relation of real dielectric constant ( $\epsilon'$ ) according to the following equation [16]:

$$\epsilon' = n^2 - k^2 = \epsilon_\infty - \frac{e^2}{4\pi^2 c^2 \epsilon_0} \left(\frac{N}{m^*}\right) \lambda^2 \quad (3)$$

where  $n$  is the refractive index,  $k$  is the extinction coefficient,  $\epsilon_\infty$  is the high frequency permittivity,  $e$  is the electron charge,  $c$  is the light velocity,  $\epsilon_0$  is the permittivity of free space,  $m^*$  is the effective mass ( $m^* = 0.26 m_0$  [27]) and  $\lambda$  is the incident wavelength. The obtained values of  $N$  were plotted in Fig.4. It is clear that, the free carrier concentration decreases with increasing the value of Sb-content. Since, the un-doped  $\text{SnO}_2$  represents the maximum free carrier concentration value of  $2.3 \times 10^{20} \text{ cm}^{-3}$ . At high concentration of Sb ( $x=0.8$ ), the free carrier starts to increase. The decrease in  $N$  can be explained due to the appearance of  $\text{Sb}_2\text{O}_3$  ( $\text{Sb}^{+3}$ ) in X-ray diffraction as shown in Fig.1-a. Since,  $\text{Sb}^{+3}$  acts as an acceptor which accept the free electrons from the conduction band. In addition, the plasma frequency  $\omega_p$  is directly proportion to the free carrier concentration as shown from the same figure (Fig.4). The values of plasma frequency were determined from the equation [16]:

$$\omega_p = \left(\frac{N e^2}{\epsilon_0 \epsilon_\infty m^*}\right)^{1/2} \quad (4)$$

The other optical parameters namely; refractive index and extinction coefficient were determined from the optical measurements in the visible region and listed in Table 1. The decreasing in  $k$  values with increasing Sb-content is expected therefore the transmittance increased with Sb-content. On the other hand, the refractive index initially decreases with increasing Sb-content and then starts to increase with further increase in Sb-content. This may be due to the behaviour of packing density as explained in ref.28.

Table 1. Electrical resistivity ( $\rho$ ) at room temperature, extinction coefficient in the visible region ( $k_{\text{VIS}}$ ) and refractive index in the visible region ( $n_{\text{VIS}}$ ) of as-deposited  $\text{SnO}_2_{100-x}\text{Sb}_x$  films of different Sb- contents.

x	0.0	1	3	5	8
$\rho$ ( $\Omega$ cm)	0.12	$1.1 \times 10^3$	$3.2 \times 10^3$	$4.4 \times 10^3$	$4.6 \times 10^3$
$k_{\text{VIS}}$	2.12	0.74	0.72	0.32	0.53
$n_{\text{VIS}}$	4.4	3.13	2.91	3.12	3.9

### 3.3. Electrical properties of as-deposited films

Results of the electrical resistivity ( $\rho$ ) measured at room temperature as a function of Sb-content of as-deposited films are listed in table 1. The presents films are resistive a matter that can be attributed to their higher degree of amorphization and the decrease in free carrier concentrations with increasing Sb-content as shown in Figs 1-a and 4, respectively. The relatively low resistivity

( $12 \times 10^{-2} \Omega \text{ cm}$ ) observed for the un-doped films is attributed to the deviation from stoichiometry due to oxygen vacancies [29, 30], which act as electron donors and increase the free carrier concentration (see Fig. 4).

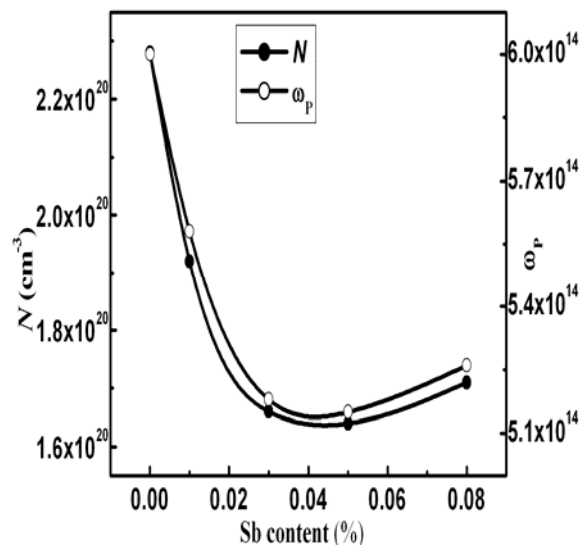


Fig. 4. Variation of free carrier concentration ( $N$ ) and plasma frequency ( $\omega_p$ ) with Sb-content of as-deposited  $\text{SnO}_{2(100-x)}\text{Sb}_x$  ( $x = 0, 1, 3, 5,$  and  $8 \text{ wt\%}$ ) films.

### 3.4. Optical and electrical properties of annealed films

To enhance the optical and electrical properties of as-deposited  $\text{SnO}_2:\text{Sb}$  films, the effect of heat treatment (annealing) was taken into account. The present study sheds light upon  $\text{SnO}_{2(100-x)}\text{Sb}_x$  films of  $x=3$  that exhibit the optimal optoelectronic properties as will seen in Fig.6.

Fig.5. shows the dependence of both electrical resistivity ( $\rho$ ) and free carrier concentration ( $N$ ) upon annealing temperature. It is clear that, the resistivity decreases with increasing annealing temperature up to  $400^\circ\text{C}$  reaching its minimum value of  $6.6 \times 10^{-3} \Omega \text{ cm}$ . Further increase of annealing temperature resulted in a significant increase of resistivity. On the other hand, the carrier concentrations of these films behave in reverse way of the resistivity. Therefore, the slight increase in carrier concentrations upon annealing ( $0-400^\circ\text{C}$ ) may be attributed to two factors i) the oxygen desorption process. Since, the desorbed oxygen would leave the free electrons behind in the film, leading to increasing of free carrier concentrations. ii) the oxidizing of the films particular the annealing process was done in the air. Beside, the abrupt decrease in carrier concentrations that observed for annealed films above temperature  $400^\circ\text{C}$  may be due to the chemisorbed oxygen and the formation of grain boundaries acting as trap sites for free electrons. Kim et al [31] and Shanthy et al [32] were obtain a similar results.

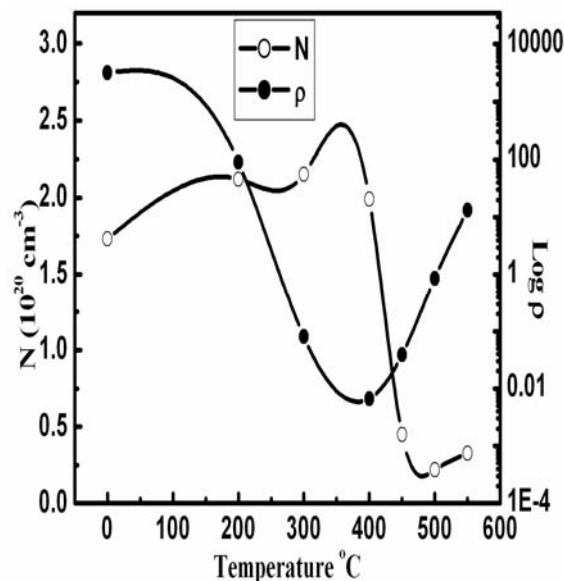


Fig. 5. Variations of free carrier concentrations ( $N$ ) and electrical resistivity ( $\rho$ ) with temperature of as-deposited and annealed  $\text{SnO}_{2(100-x)}\text{Sb}_x$  films of  $x=3$ .

Fig.6-a shows the transmittance at wavelength  $550 \text{ nm}$  as a function of temperature of  $\text{SnO}_{2(100-x)}\text{Sb}_x$  ( $x = 0, 1, 3, 5,$  and  $8 \text{ wt\%}$ ) films. It is observed that, with increase the temperature up to  $400^\circ\text{C}$  the transmittance decrease for all films with different Sb ratios. Beside, the maximum value of transmittance does not exceed  $50 \%$  for the films of  $x=0.05$ . A significant enhancement of transmittance is observed above temperature  $400^\circ\text{C}$ . Thus, the transmittance exceeds  $84$  at  $600 \text{ nm}$  for  $\text{SnO}_{2(100-x)}\text{Sb}_x$  films of  $x=3$ . The decrease in transmittance upon annealing in the temperature range from as-deposited to  $400^\circ\text{C}$  may be due to the increase in surface roughness. Therefore, the surface mobility of constituent elements increases and growth of face grains are preferred and degree of surface roughness increases due to heating. Other workers obtain the similar results [16, 33, 34]. Beside, the increase in free carrier concentration upon annealing up to  $400^\circ\text{C}$  as shown in Fig.5 leads to increase the free carrier absorption and hence degrades the transparency of these films [35, 36]. On the other hand, the significant increase in transmittance at high temperature is expected due to increase the optical energy gap resulting from the improvement of the film ordering as explained by x-ray analysis as shown in Fig 1-b. The data of optical energy gap and Urbach energy, for  $\text{SnO}_{2(100-x)}\text{Sb}_x$  films of  $x=3$ , that listed in Table 2 confirm our above discussion. Fig. 6-b. represents the transmittance spectra of as-deposited and annealed  $\text{SnO}_{2(100-x)}\text{Sb}_x$  films of  $x=3$ . This figure shows a similar behaviour to Fig.6-a. The corresponding optical energy gap of these films were calculated and listed in table 2. Although, the transmittance decreases upon annealing in the temperature range from as-deposited to  $400^\circ\text{C}$ , the energy gap increases. This increase in energy gap may be due to Burstein-Moss effect [37, 38],

particularly, the free carrier concentrations increased in this temperature range. At high temperature ( $> 400$  °C), both the transmittance and energy gap are increased.

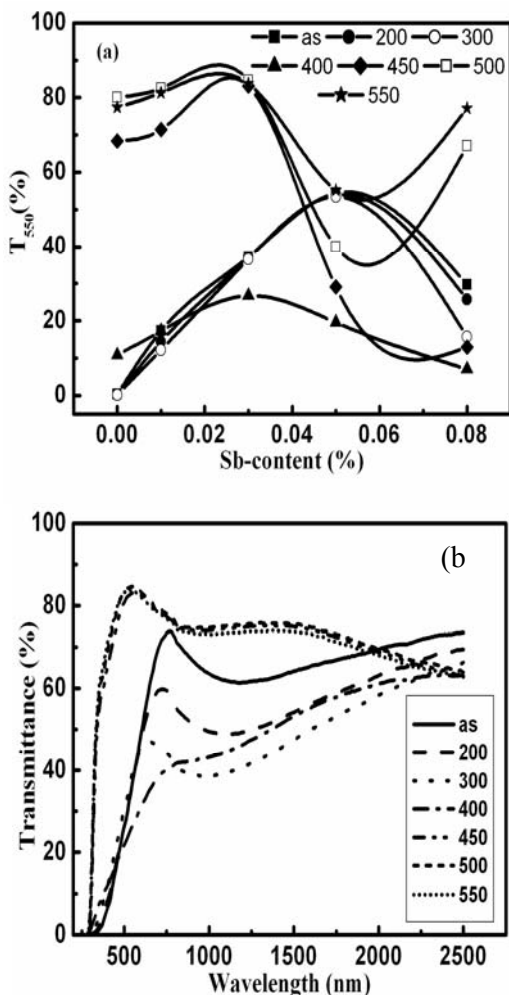


Fig. 6. Transmittance spectra a) at wavelength 550 nm ( $T_{550}$ ) as a function of Sb-content of as-deposited and annealed  $\text{SnO}_{2_{100-x}\text{Sb}_x}$  ( $x = 0, 1, 3, 5,$  and  $8$  wt%) films and b) of as-deposited and annealed  $\text{SnO}_{2_{100-x}\text{Sb}_x}$  films of  $x = 3$  in the wavelength range from 200 to 2500 nm.

Table 2. Variation of refractive index in the visible region ( $n_{\text{VIS}}$ ), optical energy gap ( $E_g$ ) and Urbach energy ( $E_U$ ) with temperature of  $\text{SnO}_{2_{100-x}\text{Sb}_x}$  ( $x = 3$  wt%) films.

T °C	$n_{\text{VIS}}$	$E_g$ (eV)	$E_U$ (eV)
0	2.91	3.06	0.749
200	2	3.07	0.702
300	1.96	3.12	-
400	1.73	2.79	-
450	2.15	3.44	0.631
500	2.11	3.54	0.628
550	2.12	3.52	0.602

Fig.7. represents the variations in refractive index ( $n$ ) of the as-deposited and annealed  $\text{SnO}_{2_{100-x}\text{Sb}_x}$  films ( $x=3$ ) with wavelength. The average refractive index values in the visible region ( $n_{\text{VIS}}$ ) were calculated and listed in table 2. It is clear that, the refractive index decreases with annealing temperature up to 400 °C and then starts to increase with further increase in the temperature. It is known that [23, 39] the behaviour of refractive index is due to reflection (R %). Therefore, the dependence of reflection on wavelength at different temperatures is shown in Fig.8.

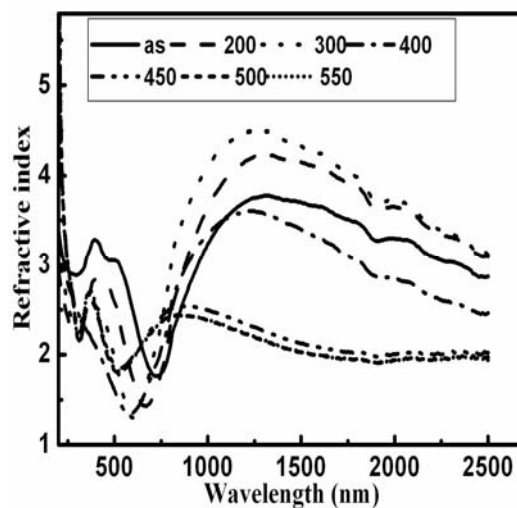


Fig. 7. Variation of refractive index with wavelength of as-deposited and annealed  $\text{SnO}_{2_{100-x}\text{Sb}_x}$  films of  $x=3$ .

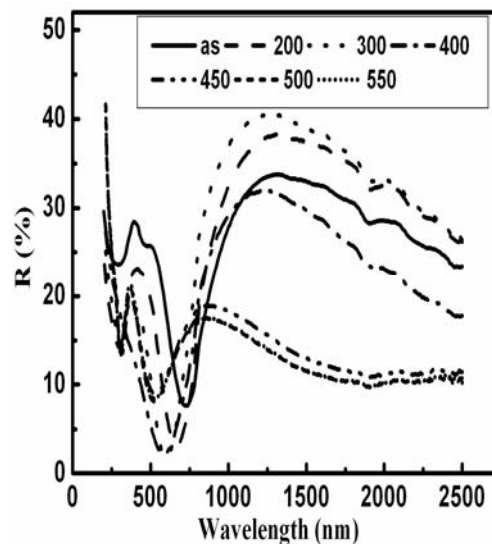


Fig. 8. Reflection spectra (R %) of as-deposited and annealed  $\text{SnO}_{2_{100-x}\text{Sb}_x}$  films of  $x=3$  in the wavelength range from 200 to 2500 nm.

#### 4. Conclusions

The e-beam evaporation technique was used to prepare transparent conducting SnO<sub>2</sub>:Sb films. The effect of Sb-content and annealing temperature was taken into consideration. The as-deposited un-doped SnO<sub>2</sub> films showed low transmittance and low electrical resistivity associated with amorphous structure. With increasing the Sb-content, both the transmittance and resistivity increased. The SnO<sub>2</sub><sub>100-x</sub>Sb<sub>x</sub> films of x=3 were selected to study the effect of annealing temperature. It was found that, the transmittance at wavelength 550 nm was significantly increased up to 84 % at temperature 450 °C and these films represented direct allowed transition with optical gap 3.06 to 3.54 eV. The temperature above 400 °C was sufficient to enhance the structure of these films. The low resistivity of  $6 \times 10^{-3} \Omega$  was recorded at the same temperature. It was found that, the refractive index, extinction coefficient, free carrier concentrations, plasma frequency and Urbach energy were very sensitive to Sb-content and annealing temperature. The optimized low resistivity at room temperature and high transmittance at wavelength 550 nm, as well as chemically stable up to high temperatures for the present films, may suggest their suitability for many applications in the field of optoelectronic technology.

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