Journal of Engineering and Applied Sciences 13 (23): 9838-9843, 2018
ISSN: 1816-949X

Effect Annealing and Irradiation on Structural-Optical Properties of Tin Oxide Thin Films

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Abstract: Tin Oxide (SnO_2) thin films were used in many applied such as in solar cells, optoelectronic devices, heat mirror, gas sensors, etc. The thermal evaporation technique was used for the synthesis of the tin oxide (SnO_2) thin films. The structural properties were analyzed by Fourier Transforms Infrared spectroscopy (FTIR). These results shown vary of bonds, shapesandpositions of the peaks. When annealing temperatures and affected by irradiation were increased. Optical properties of SnO_2 thin films were investigated using the UV-VIS (T70/80) spectrophotometer. The highest transmittance spectra with annealing and irradiation while the lowest transmittance spectra at annealing temperature 623 K. The value of refractive index n was decreased after annealing temperatures and irradiation. The extinction of coefficient k was found to be very small. The energy gap was decreased with annealing temperature but the energy gap for the sample annealed at 623 K was increased after irradiation.

Key words: FTIR spectrum, SnO_2 thin films, annealing temperatures, irradiation, optical properties, energy

INTRODUCTION

One of the more important and attractive research topics in physics and material sciences oxides thin film materials (Agbo, 2012). Thin films (SnO_2) are used in some critical applications as transparent conductors, for example, electrodes in solar cells covering the front surface of these devices or electrodes in organic semiconductors based devices, electromagnetic shielding maintaining transparency, flat-panel displays, defrosting windows, low-emissivity windows and oven windows (Mahdi, 2012). Ionizing radiations like (X-rays, gamma-rays, beta particles, alpha particles, fission fragments, etc.) are existent in many fields that contain industry, medicine, military, particle accelerator-based research, nuclear power plants, etc. It is very necessary to guarantee that the radiation levels in the environment surrounding these fields are within the permissible limit which can be determined by proper dosimeters (Antar, 2014). Depending effect of ionizing radiations on metal oxide thin films both the radiation dose and the film thickness refer to the parameters of the thin films. It is previously an established fact that the degradation is caused by higher radiation doses and thinner films (Elhaleem, 2013). When solid material exposure to ionizing radiations produces interaction of radiant energy with matter lead to changes in the microstructure properties of the material which in turn affects the optical, electrical and other physical properties of the material (Al-Hamdani *et al*., 2014). The present research reports our studies on the effect of different radiation on structural-optical properties of SnO_2 thin films prepared by using thermal evaporation method.

MATERIALS AND METHODS

Experimental work

Preparation tin oxide (SnO_2) thin film: Thin films of tin oxide transparent conductive are prepared by using thermal evaporation technique (Edwards system) at room temperature under a low vacuum pressure of 10^{-6}Torr. Spectroscopically pure (about 99.9%) tin oxide powder is used as the source material. Glass slides have been used as the substrates after cleaned by alcohol with ultrasonic waves to improve the adhesion of the coating to the glass substrate. Films of thickness 500 nm are formed through evaporations process.The resulting films then passed through a tube furnace for different annealing temperatures of 623,723 and 823 K through annealing time duration 2 h/sec for each ether (Ikhmayies, 2012).

Irradiation substrate tin oxide (SnO_2) thin film: The irradiated sources have been installed at a distance 1 cm above the annealing substrate (SnO_2) thin film at 623 K-30 sec for each user irradiated sources to research. All information for various irradiated sources listed in the Table 1.

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Table 1: Information for various irradiated sources

Isotope	Activity	Date of manufacture	$T_{1/2}$	E(keV)	I%	Notes
$^{90}Sr_{52}$	3 μCi	1978	28.79Y	546.00	I_β	Manmade (fission), Q [β(100%) = 546.00 keV
$^{60}Co_{33}$	1 μCi	2009	5.27Y	1173.24	$I_\gamma = 99.97$	Manmade (neutron activation) Q [β (100%) = 2823.9 keV
				1332.50	$I_\gamma = 99.99$	
				318.13	$I_\beta = 99.93$	
$^{241}Am_{146}$	9 μCi	1976	432.2Y	59.54	$I_\gamma = 35.9$	Manmade (neutron activation) Q [α (100%) = 5637.8
				5485.6	$I_\alpha = 84.5$	keV+S.F
				5442.8	$I_\alpha = 13.0$	

RESULTS AND DISCUSSION

Fourier transformation infrared results: The samples were examined by Fourier Transmission Infrared spectroscopy (FTIR; Bruker-Equinox 55; Bruker Optics, Billerica, MA) is taken in the frequency range (400-4000) cm^{-1} in transmission mode. FTIR results give information about phase composition and the way in which oxygen is bound to metal ions. Figure 1 shows that the FTIR spectrum of (SnO_2) thin films prepared by evaporation methods for varies annealing temperatures. The absorption bands of (SnO_2) at 623 K, shown in Fig. 1a the bands at (489.75) cm^{-1} refer to stretching vibrations Sn-O-Sn, (837.35) cm^{-1} refer to metal hydroxide Sn-OH and 1087.83 cm^{-1} refer to O-H stretching vibration (Sharma *et al*., 2007; Sagadevan and Podder, 2015). During annealing temperature 723 K for (SnO_2) thin film, the absorption bands changing shown in Fig. 1b the new absorption bands appearance at (415.30, 433.31, 513.56, 543.05, 600.16, 688.30 and 784.86) cm^{-1} refer to the characteristic ant symmetric stretching vibrations of Sn-O-Sn (Petrov *et al*., 2012; Khan *et al*., 2010). The band (827.77) cm^{-1} refers to metal hydroxide networking Sn-OH in the thin films and the peaks at (1066.20, 1125.96, 1990.83 and 2015.14) cm^{-1} were due to the vibration mode of (O-H) group. So, the absorption bands appeared and disappeared off the (SnO_2) thin films after annealed at 823 K shown in Fig. 1c three absorption bands appearance at (417.76, 437.75 and 520.96) and shift two bands at (488.15, 830.23) cm^{-1} were referred to chemical bonding Sn-O, Sn-O-Sn vibration stretching (Sagadevan and Podder, 2015). The peaks at (1083.96, 1998.98 and 2340.88) cm^{-1} were assigned to the vibration mode of O-H group (Sharma *et al*., 2007). The absorption bands of tin oxide (SnO_2) thin film annealing at 623 K and irradiation are shown in Fig. 1d. The absorption bands at (408.65, 430.98, 474.26, 578.12, 600.28, 655.52 and 725.54) cm^{-1} can be assigned to O-Sn-O, Sn-O-Sn stretching vibrations and to lattice vibrations, respectively (Khan *et al*., 2010). The absorption bands at (1361.92, 1419.44, 1508.89 and 1583.41) cm^{-1} were assigned as the vibration mode of O-H group, the peaks at (827.27, 883.84 and 1605.02) cm^{1} refer to Sn-OH (Sharma *et al*., 2007).

Optical poroperties: Tin oxide (SnO_2) thin films deposited onto the glass substrate at room temperature. The transmittance and the absorbance spectra measurements were done between (300-900) nm. Which that measured by used the instrument (T70/T80 UV-Visible spectrophotometer).

The optical transmittance spectrum: The thin films were treated for annealing at different temperatures and expose irradiation by different sources illustrated in Fig. 2. The transmittance for tin oxide (SnO_2) thin films in the visible light region (300-900 nm) and from the diagram, tin oxide (SnO_2) thin film annealed at 623 K exhibits transmittance in the (370-900 nm) which maximum transmittance range is 40-50% where the (SnO_2) films annealed at 723 K shows high transmittance above 40%. The film annealed at 823 K initially shows the lowest transmittance and gradually increases this is in a good agreement with the results obtained by Baco *et al*. (2012) while its highest transmittance 90% for annealing (SnO_2) thin film at 623 K with irradiation. Nevertheless, the transmittance for all annealed samples rises with increasing wavelength and this also, approved by Singh *et al*. (2013). The average transmittance of annealing films is <70% for annealing with irradiation films, there is an increment of the transmittance when absorb the expose irradiation for annealing films.

Optical constants; refractive index n and extinction coefficient k: The refractive index n and the extinction coefficient k are the most important optical parameters of tin oxide (SnO_2) thin films which are calculated from transmission and reflectance spectra. The refractive index n can be obtained with the following Eq. 1:

$$n = \left[\left(\frac{1+R}{1-R}\right) - \left(K^2 - 1\right)\right]^{1/2} + \frac{1+R}{1-R} \quad (1)$$

n is the refractive index for the refracted index for thin film and R is the reflectance. k is represent extinction coefficient which is the definition as that amount loosed in energy because of reaction between the light and the medium charge Can be calculating excitation coefficient k for thin films by used Eq. 2 (Sagadevan and Podder, 2015):

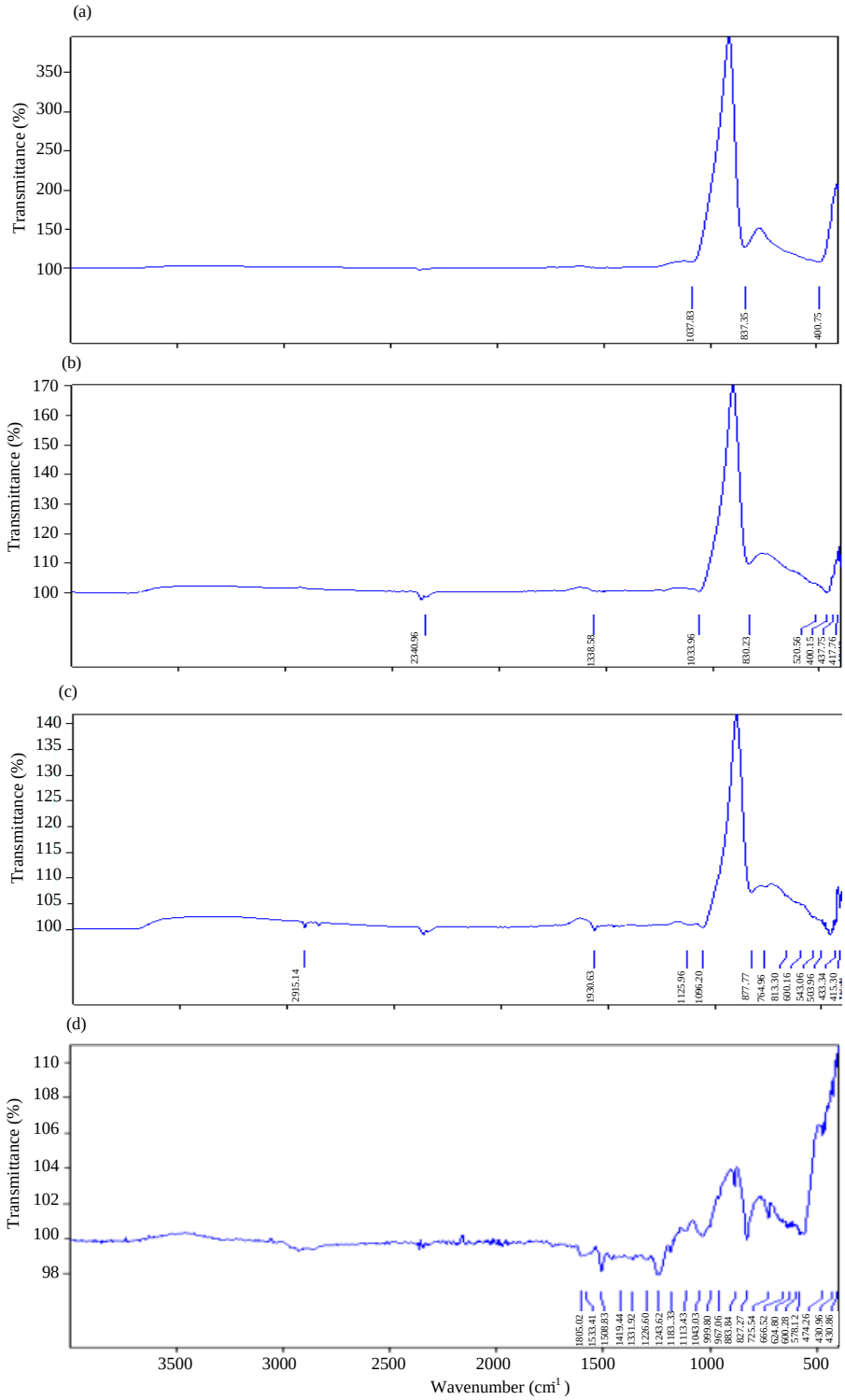

Fig. 1: FTIR spectrum of the SnO_2 thin films: a) Annealing at 623 K; b) Annealing at 723 K; c) Annealing at 823 K and d) Annealing at 623 K with irradiation

$$K = \frac{\alpha\lambda}{4\pi} \quad (2)$$

Where:

α = The absorption coefficient

λ = The excitation wavelength which that used for the pumped thin film

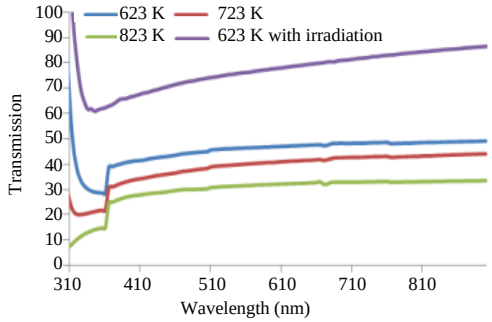

Fig. 2: Optical transmittance for tin oxide (SnO_2) thin films at different annealing temperatures and irradiation

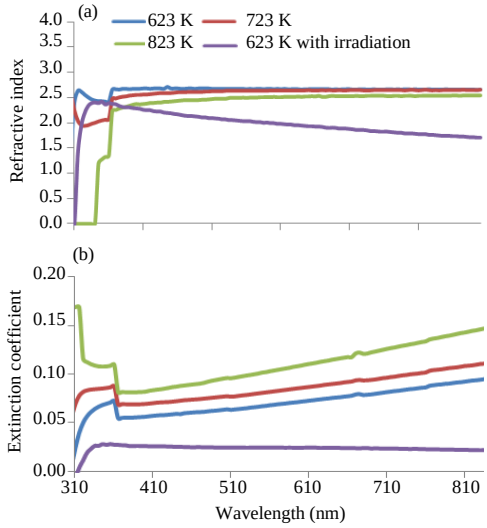

Fig. 3: Tin oxide (SnO_2) thin films annealed at different temperature and irradiation; a) Dispersion of refractive index n and b) Dispersion of extinction coefficient k with wavelength

Figure 3 shows the variation of refractive index and the extinction coefficient with wavelength and this amount similar behavior also found by Singh *et al.* (2013). It can be seen that the refractive index n decreased for tin oxide (SnO_2) thin films with increase annealing temperatures and when irradiation by several sources for annealing (SnO_2) thin film at 623 K. Figure 3a shows the values of refractive index n for (SnO_2) thin films were (2.7, 2.6, 2.4 and 2.3) at λ = 410 nm in different annealing temperatures (623 K, 723 K, 823 K and 623 K with irradiation), respectively.

The extinction coefficient k increased for tin oxide (SnO_2) thin films with increase annealing temperatures and decreased with irradiation for annealing thin film at 623 K. Fig. 3b shows the values of extinction coefficient k for (SnO_2) thin films were (0.06, 0.081, 0.083 and 0.025) at

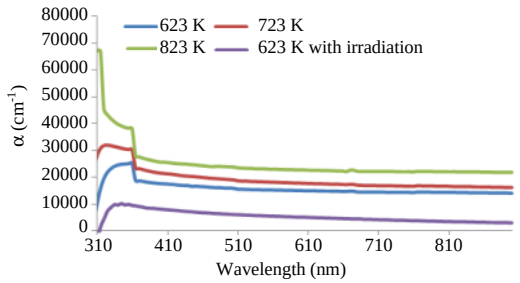

Fig. 4: Optical absorption coefficient α as wavelength for tin oxide (SnO_2) thin films at different annealing temperature and irradiation

λ = 410 nm in different annealing temperatures (623 K, 723 K, 823 K and 623 K with irradiation), respectively. These results point to oxidation and diffusion of tin atoms from the deposited thin films on the glass substrate.

Absorption coefficient α: The intensity of falling light (I_0) is greater than the intensity of emerging light when the light passes through a media specific and the reason to do the weaken intensity of falling light is the reflection in the limits surface between them or because of desperation suffered by the beam because of matter in the media or direct absorbed by the matter. If the light with intensity I_0 incident on the thin films with thickness *x*, transmitted intensity I, the expressed the Lambert-Beer Law as Eq. 3 (Kumar *et al.*, 2009):

$$I = I_0 e^{-\alpha x} \quad (3)$$

The absorption coefficient α (cm^{-1}) can be calculated from the lambert's formula according to the following Eq. 4:

$$\alpha = 2.303 \log \frac{I}{I_0} \quad (4)$$

Figure 4 shows the absorption coefficient α as a function of wavelength λ. The absorption edge shifted and broadened to low wavelength. At high energy (very low wavelength) absorption coefficient α is observed ($\alpha > 10^4$ cm^{-1}) due to the domination of fundamental band-gap, the high values of absorption coefficient owing to direct electronic transitions of the energy and momentum of the electron and photon, even as low energies (for higher wavelength) absorption coefficient α is observed ($\alpha < 10^4$ cm^{-1}), the low values of absorption coefficient due to indirect electronic transitions of the energy and momentum of the electron and photon by phonon helps (Tauc *et al.*, 1966). Based on the diagram, the absorption coefficient values for (SnO_2) thin films in

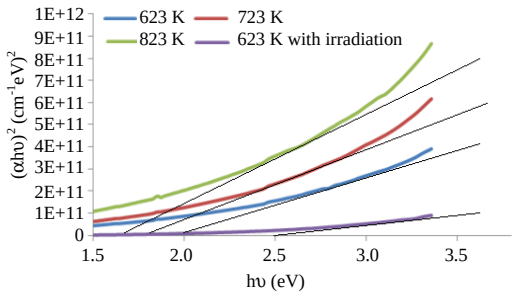

Fig. 5: Plot of $(h\alpha)^n$ against photon energy for tin oxide SnO_2 films annealed at different temperature and irradiation

this study are in the range of 10^4-10^5, so that, electronic transitions are the direct electronic transitions that similar to the previous study (Buchholz *et al*., 2012).

Optical Energy gap E_g: The absorption of photon energy due to excitation of the electron from the valence band to conduction band can occur in two ways usually either in direct or indirect transitions. These transitions are described by Tauc relation (Balkanski, 1992):

$$\alpha h\upsilon = B\left(h\upsilon - E_g\right)^r \quad (5)$$

Where:

hv = The photon energy
B = Constant and has different values for different transitions
E_g = The energy gap
r = Exponential constant

Its value depended on the material and type of optical transition where r = 1/2 for allowed direct transition, r = 3/2 for the forbidden direct transition, r = 2 for allowed indirect transition and r = 3 for the forbidden indirect transition. In order to determine the optical energy band gap of thin films, the diagram of $(hv\alpha)^n$ versus hv has been plotted by replacing n = 1/r which are n = 2, 2/3, 1/2 and 1/3 (Stenzel, 2005).

In this study, the value of n was chosen to be 2 (for allowed direct transition band gap) as illustrated in Fig. 5 because it was the most probable transition and the diagram gave the best line fit. The energy gap was found that decreased with annealing temperature and increases at annealing with irradiation. The highest energy gap was obtained from the sample annealed at 623 K with irradiation at 2.5 eV while the lowest energy gap was from the sample annealed at 1.8 eV and 1.7 eV for 723 K and 823 K, respectively. Singh *et al.* (2013) illustrated the decrease in energy gapvalue with increasing annealing temperature

Table 2: Energy gap for Table: energy gap for tin oxide SnO_2 thin films with different annealing temperature and irradiated

Tin oxide SnO_2 thin films with different annealing temperatures (Energy gap E_g(eV))			
623 K	723 K	823 K	623 K with irradiation
2	1.8	1.7	2.5

because that eliminating a part of the oxygen vacancies and localizes the oxygen atoms at interstitials by the annealing process. The induced oxygen interstitials form separate band defects in the band gap region. While the increase in the optical band gap when using irradiation with annealing temperature due to increasing carrier concentration is related to the rise of the Fermi level in the conduction band of a degenerate semiconductor such as that found in some degenerate semiconductors and is known as a Moss-Burstein shift (Kharisov *et al*., 2013). The difference of direct band gap with annealing temperature and irradiation was obtained in Table 2.

CONCLUSION

Tin oxide (SnO_2) thin films were prepared by thermal evaporation on glass substrates at room temperatures. The effects of annealing temperatures and irradiation on the structural and optical properties of SnO_2 thin films were studied. The character of FTIR spectra and positions of the bands have been shown to vary with the different annealing temperatures at the constant time. The mean kinetic energy of the tin atoms increased through the increase annealing temperatures. The chemical bondings like (Sn-O) vibrating and (O-Sn-O) stretching modes are formed, respectively. The irradiated have been proven to change the shape of FTIR spectra and positions of the peaks resulting in the degradation of their performance. The UV-VIS spectrophotometer showed that highest transmittance 90% for annealing (SnO_2) thin film at 623 K with irradiation. The average transmittance for (SnO_2) thin films in different annealing temperatures is <70%. The values of refractive index n were (2.7, 2.6 and 2.4) at λ = 410 nm for annealing (SnO_2) thin films (623 K, 723 K and 823 K) respectively while decrease to 2.3 for annealing thin film 623 K after irradiation. The extinction coefficient k for (SnO_2) thin films was found very small with annealing temperatures and irradiation. The energy gap was 1.7 eV obtained from the sample annealed at 823 K while the energy gap was 2.5 eV for the sample annealed at 623 K with irradiation.

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