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Gamma irradiation study of tin oxide thin films for dosimetric applications

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Electrical and optical properties of as-grown and gamma irradiated films of tin oxide, grown on glass substrate by electron beam evaporation at a substrate temperature of 250 °C have been studied. The as-grown films show conductivity ranging from ~0.02 to ~1 S-cm^{-1} and the irradiated films show the increment in conductivity value upto ~1.18 S-cm^{-1}. The optical band gap of films has been determined from the transmittance and reflectance spectra in the wavelength range of 400-850 nm at room temperature. The optical band gap of as-grown tin oxide thin film is direct band gap and has a value of ~2.85 eV. Blue shift has been observed in the band gap when exposed to 60 Gy cumulative dose of gamma radiation with a value of ~2.76 eV. XRD analysis has shown that the as-grown films were polycrystalline in nature having crystallite size of 33 nm and there is an improvement in crystallinity and grain size after gamma exposure. This work demonstrates that tin oxide thin films are sensitive to gamma radiation and it can be used for dosimetric applications.

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Keywords: Gamma radiation, Tin oxide, Thin films, Oxygen vacancies (V_O), Electron beam evaporation

1. Introduction

Silicon based semiconductor devices have been used extensively over the past few decades for various applications including radiation sensors. Over the past three decades, more emphasis has been given to detect high energy radiations such as gamma and X-rays for national security, environmental safety, military applications, biology, medicine and basic sciences. Research interest has been generated to look out new metal oxides in single layers or multi layers form for ionising radiation interaction [1]. The reduction in device dimensions has led to the need for alternative high dielectric constant (k) oxides to replace silicon-di-oxide as the gate dielectric in MOS devices [2]. Tin oxide is IV–VI semiconductor material that can be used for a variety of applications such as electronic displays, thin film solar cells, gas sensors, infrared-reflecting glass windows, anti-reflection coatings, transparent electrodes and anti-static sheets. Considering these applications, behaviour of tin oxide under the harsh conditions such as gamma radiation environment is an important aspect which needs some attention. Ionizing radiation changes the physical properties of the materials they penetrate. The changes are strongly dependent on the internal structure of the absorbed substances. It is known that ionizing radiation causes structural defects called color centers or oxygen vacancies in oxides leading to their density change on the exposure of gamma rays [3]. The influence of radiation depends on both the dose and the parameters of the films including their thickness however, the degradation is more sever for higher doses. A number of efforts have been devoted to investigate the influence of gamma radiation on the properties of metal oxide materials/thin films, e.g. In_2O_3, CeO_2, NiO, TeO_2, SiO [4-13]. It is necessary to investigate the changes in the properties of tin oxide thin film structures under the influence of gamma radiations. Very limited literature is available on the gamma radiation effect on Al/SnO_2/Si (MOS) structure [14,15], no efforts have been made so far on electrical and optical properties of tin oxide thin film under gamma environment. In this work we report the effect of gamma irradiation on tin oxide thin films. The study will show the applicability of tin oxide thin films for high dose gamma dosimetry.

2. Experimental

Tin oxide thin films have been grown on glass substrates by electron beam evaporation from a high quality commercially available SnO_2 powder (Sigma-Aldrich) with 99.99% purity using graphite boat in a high vacuum of 2×10^{-5} mbar and the substrate temperature at 250 °C. Prior to deposition, the glass substrates (Size: $7.5 \times 2.5 \times 1.35$ cm^3) have been cleaned with soap water, NaOH solution and acetone and then rinsed by ultrasonic vibration in deionised water and dried. Preceding each cleaning step, the substrates have been rinsed thoroughly in deionised water of resistivity of 10 M Ω-cm. The distance between substrate and source has been kept at 15 cm. The quartz crystal thickness monitor has been used for monitoring the film thickness and deposition rate during evaporation. Tin oxide films having the thickness of 500 nm have been deposited at a deposition rate of ~1.5 Å/s.

An array of ^{60}Co gamma radiation source (Gamma Chamber-900) has been used for exposing the thin film samples to gamma radiation at room temperature. A total dose of 60 Gy in steps of 20 Gy has been given by the gamma photons at a dose rate of 2 Gy/min. Electrical measurements of the film have been done by two-probe method. Ohmic contact has been made on film using high-purity silver paste and fine copper single wires. I-V characteristics measurements have been done by using Keithley Electrometer 6517A. A double beam UV-VIS-NIR Spectro photometer (Perkin-Elmer Lambda 900) has been used for the optical transmission (%T) and reflectance (%R) measurements in the range of 400-850 nm, at room temperature. Philips X-ray diffractometer using Fe (λ = 1.936 Å) X-ray source has been used for structural analysis.

3. Results and discussion

Fig. 1 shows X-ray diffractograms of the electron beam evaporated tin oxide thin film before and after gamma irradiation. The peaks identified comparing with JCPDS file for SnO corresponds to (001), (101), (002), (112), plane of reflection for tetragonal type of SnO. It is observed from the Fig. 1 that the film has grown predominentaly in (101) direction. Banerjee and Das [16] have shown that tin oxide films grown at 350 °C without added oxygen or with low P(O_2) exhibit a crystalline SnO structure by the electron beam evaporation method. SnO structure obtained in this work shows that there is a reduction from oxidation state of Sn ion from +4 state to +2 during electron beam evaporation. It should be noted that the electron beam evaporation technique decomposes the SnO_2 into SnO and O [17]. It is also observed that the peak intensity increases after irradiation confirming the increment in crystallinity after gamma exposure. A similar result of partial crystallization has been observed by Arshak et. al. [12] for gamma influenced In_2O_3/SiO structure. Assuming a homogeneous strain across crystallites, the crystallite size of micro-crystallite can be calculated from the full width half maximum (FWHM) values by using the well known Scherrer formula [18] for crystallite size broadening of diffraction peaks,

$$D = 0.94\,\lambda / \Delta_{2\theta} \cos\theta \qquad (1)$$

where D is crystallite size, λ is the wavelength of X-ray, Δ is the full width half maximum (FWHM) of diffraction peak and θ is the diffraction angle. The crystallite size of the tin oxide thin film of 500 nm thickness comes out to be ~33 nm and ~50 nm before and after gamma irradiation, respectively.

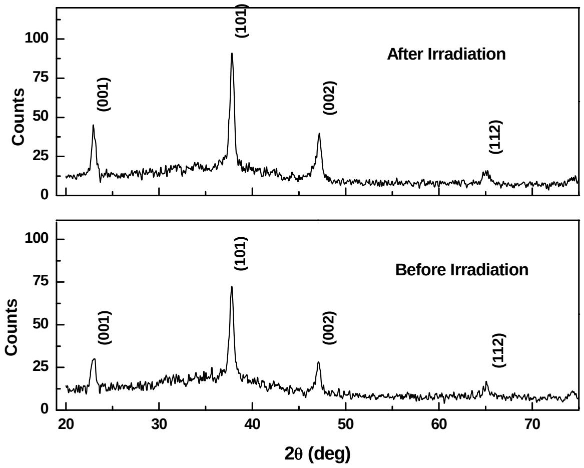

Fig. 1. X-ray diffractogram of tin oxide thin film before and after gamma irradiation.

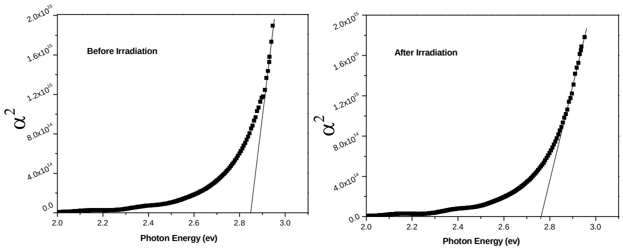

Fig. 2. α^2 vs photon enery(hv) of tin oxide thin film before and after gamma irradiation.

Fig. 2 shows the plots of α^2 (α is optical absorption coefficient) versus photon energy hν (wavelength range 400-850 nm) obtained at room temperature for tin oxide thin film before and after gamma irradiation. The absorption coefficient α has been calculated from the Mott's & Davis model [19] for the direct allowed transition by the equation,

$$\alpha(\nu)h\nu = \beta(h\nu - E_{Opt})^{1/2} \qquad (2)$$

where α is the absorption coefficient, E_{Opt} is the optical band gap, hν is the energy of the incident photon and β is a constant. The value of the direct optical band gap Eg has been determined by extrapolating the linear region of the plots to zero absorption and is 2.85 eV. It should be noted that band gap of SnO is lying somewhere in the range of 2.5–3 eV [20,21]. A decrement in Eg from 2.85 eV to 2.76 eV has been observed, when the tin oxide thin films are irradiated to a gamma cumulative dose of 60 Gy. This decrement confirms that decrease in band gap with increasing dose may be attributed to the cause that irradiation increases defects such as oxygen vacancies (V_O) in the material which may lie just below the conduction band. Arshak and Korostynska [13] have shown that optical band gap for TeO_2 film has decreased from 3.75 eV to 3.45 eV with increasing radiation dose up to a level of 36 Gy. Depending on the electronic structure of the material, the nature of oxygen vacancies changes dramatically [22]. Under the rudimentary model of the charge transfer, the Fermi level in the irradiated sample shift upwards with respect to its position in the non-irradiated structure, filling the holes states in plane bands [3]. Due to the gamma irradiation band tail might have been created and E_{Opt} has decreased.

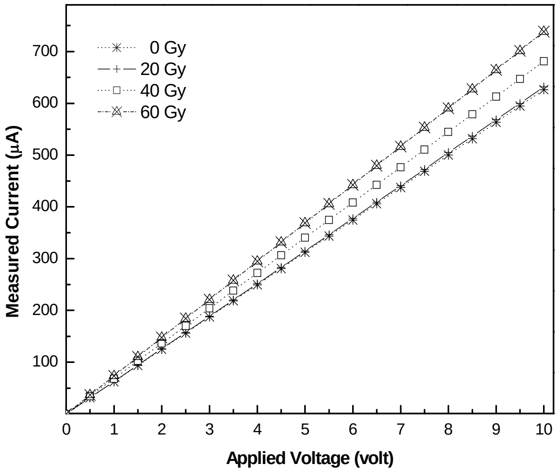

Fig. 3. Current vs applied voltage plot of tin oxide thin film at various cumulative doses.

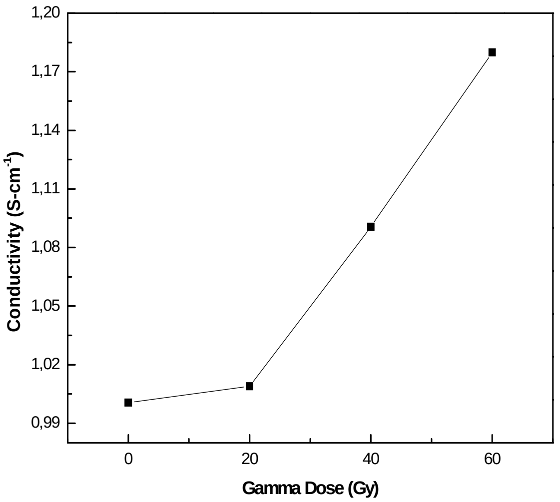

Fig. 4. Conductivity vs cumulative dose plot of tin oxide thin film at various cumulative doses.

Influence of gamma radiation on the electrical properties of tin oxide thin film has been shown in Fig. 3 and Fig. 4. Fig. 3 depicts the I-V characteristics of the tin oxide thin films measured for the applied voltages of 1 to 10 volt at various cumulative doses. The ohmic behavior of the thin film has been confirmed by the linear variation of the I-V characteristics of the tin oxide thin film before and after gamma irradiation with applied bias. Some of the published work [12, 13] on metal oxide thin and thick films also shows the same phenomenon. No appreciable change in current values has been observed at the irradiation of 20 Gy at the dose rate of 2 Gy/min. However, an increase in current has been observed at the cumulative doses of 40 Gy and 60 Gy for the same dose rate. Fig. 4 depicts the plot of conductivity versus cumulative dose of electron beam evaporated tin oxide thin film. The obtained conductivity for the as grown film was 1.00 S-cm^{-1} and has increased to the value of 1.09 S-cm^{-1}, 1.1 S-cm^{-1}, and 1.18 S-cm^{-1} for the 20 Gy, 40 GY and 60 Gy cumulative gamma doses, respectively. The defects created by the gamma photons acts as a majority carrier which plays a role in the conduction mechanism to increase the conductivity of the material.

4. Conclusions

The radiation response of electron beam evaporated tin oxide thin films of 500 nm has been investigated using ^{60}Co gamma source. From the XRD analysis it has been found that the crystallite size of the material has increased from ~33 nm to ~50 nm and the changes in atomic arrangement can be attributed for gamma dose. The obtained optical band gap value has decreased with the radiation dose. The value of leakage current and conductivity of tin oxide thin film increased considerably with the increase in radiation dose. Radiation induced changes in the electrical and optical properties of thin films resulted in the degradation of their performance, such as increase in conductivity and decrease in optical band gap has been observed in this work. We explored the possibility of tin oxide thin film for using gamma dosimetric applications beyond 20 Gy dose.

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