

Effect of gamma radiation on structural, optical and electrical properties of ZnO thin films

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Thin films of zinc oxide (ZnO) were prepared on cleaned glass substrate by a solution of zinc acetate in a carrier gas of air using spray pyrolysis technique. Then the prepared films were subjected to gamma radiation at different dose levels. Before and after gamma radiation, the optical, structural and electrical properties were studied. From UV results, it was found that the optical band gap decreases with the gamma radiation dose. XRD pattern revealed the poly crystalline nature of the sample and illustrated that the crystalline nature of the sample increases with gamma radiation dosages. Electrical studies showed the increase in current with respect to the applied gamma radiation dose.

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1. Introduction

For the last two decades, ionizing radiation has emerged as one of the important fields owing to its broad applications in several fields such as industry, medicine (cancer treatment), radio biology, military and nuclear power production [1-3]. When a solid material is exposed to ionizing radiation it undergoes changes in its structural properties. Also, the gamma ray irradiation will affect the optical and electrical properties of the material, which can be used to analyze the radiation levels. The increased use of radiation therapy in the field of medical sciences and incidents like the Japanese nuclear power station disaster, the relevance of dosimeters has gone up. In nuclear power stations, it is very important to monitor the radiation level to maintain radiation proof surrounding for the workforce. To constantly monitor radiation levels and to initiate alert in case of an emergency, novel radiation sensors are needed. These novel sensors need to be fabricated at low cost [4-8].

ZnO is one of the emerging materials owing to its excellent electrical and optical properties. The high chemical and mechanical stability along with low cost makes ZnO potentially an important material compared to indium tin oxide (ITO) and tin oxide (SnO₂). Applications include inorganic and organic solar cells, different types of sensors, light emitting diodes, heating devices and electro acoustic devices [9-11]. Generally, the spray technique is used to prepare the metal oxide based thin films. However, ZnO thin films were prepared by various methods such as sputtering, chemical bath deposition, spin coating, dip technique, spray pyrolysis and pulsed laser deposition. Among the methods, spray pyrolysis technique has the advantages like low cost, no vacuum requirement and versatility. It is also observed that this technique is very effective for large area coatings [12,13]. The present work

is aimed to study the optical, structural and electrical properties of gamma ray induced ZnO thin films.

2. Experimental techniques

The schematic diagram of the experimental setup of spray technique is shown in Fig. 1. Uncontaminated substrate surface is one of the most important parameter for the film formation. Microscopic glass slides were cleaned initially with soap water followed by rinsing in distilled water and then kept in dilute HNO₃ for 24 hours. Afterwards the substrates were rinsed with distilled water and treated with sodium hydroxide solution. These acid-alkali treated substrates were cleaned with hot isopropyl alcohol in ultrasonic bath for 40 minutes and then dried by blowing hot air. ZnO thin films were prepared from the aqueous solution of zinc acetate (0.5) dissolved in deionized water. The substrate temperature was kept at 400°C. Distance between substrate and the spray nozzle was about 20 cm. Air was used as a carrier gas and the spray rate of the solution was set as 2 ml per minute. The spray deposited films were subjected to gamma radiation. Optical properties of the films were studied using Perkin-Elmer lambda 35 spectrophotometer in the range of 200 - 2200 nm. XRD pattern of the polycrystalline ZnO films were carried out by RICH SIEFERT X-ray powder diffractometer using CuK α ($\lambda=1.5405$ Å) radiation. The sample was scanned for a range of angles (2θ) 10 – 70° at a scan rate of 1°/min. Electrical studies were done using two probe setup. In gamma ray chamber (5000) Co⁶⁰ was used as radioactive source. The dose rate was set to be ~8.4 Gy and the dose at 80 Gy. The actual gamma ray chamber and schematic diagram of the same are depicted in Fig. 2 and 3.

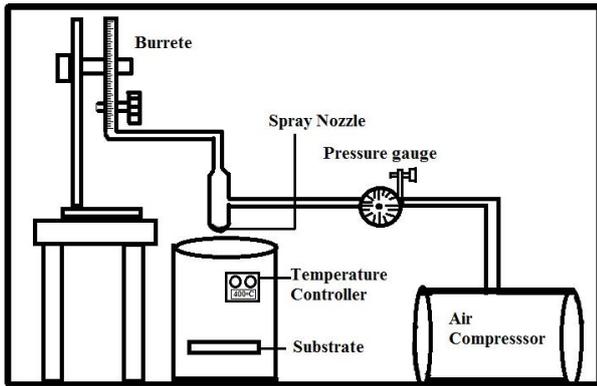


Fig. 1. Schematic diagram of Spray technique unit



Fig. 2. Gamma ray chamber

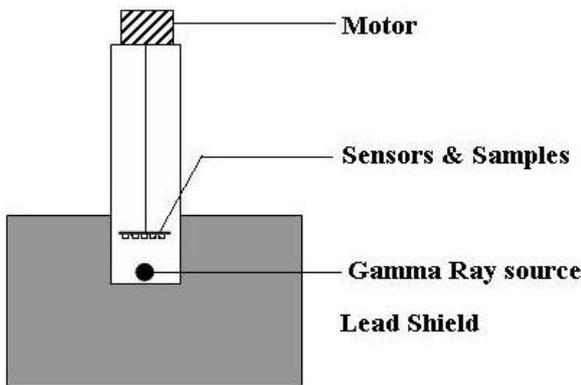


Fig. 3. Gamma ray chamber schematic diagram

3. Results and discussion

3.1. Optical studies

The optical transmittance spectra of spray deposited ZnO thin films were taken before and after gamma ray exposure and is recorded in the wavelength range of 200-2200 nm. The ZnO thin films deposited on the glass substrate yielded a transmittance of ~65 % for pure (nil

dose) and was found to increase with gamma ray dosage, which shows the improvement in transmittance [1] as depicted in Fig. 4. The optical band gap was found to decrease with the increase of gamma ray dose (Fig. 5). The variation of the optical band gap with the radiation dose is depicted in Fig. 6. This clearly shows the decrease of optical band gap from 3.56 to 3.17 eV for a gamma radiation dose of 240 Gy. Decrease in the optical band gap is mainly due to the raise in the energy width of the band tails of localized states. Even at ambient temperature, at the time of gamma irradiation there is creation of structural defects and annihilation of structural defects within the film [1,14]. Both, formation and annihilation of structural defects coexist together. At higher doses of gamma radiation, creation of structural defects will become predominant than its annihilation. The extent of structural defects created owing to irradiation [1], becomes more than the extent of structural defects annihilated. The present study clearly indicates the decrease in optical band gap with increase in radiation dose [15].

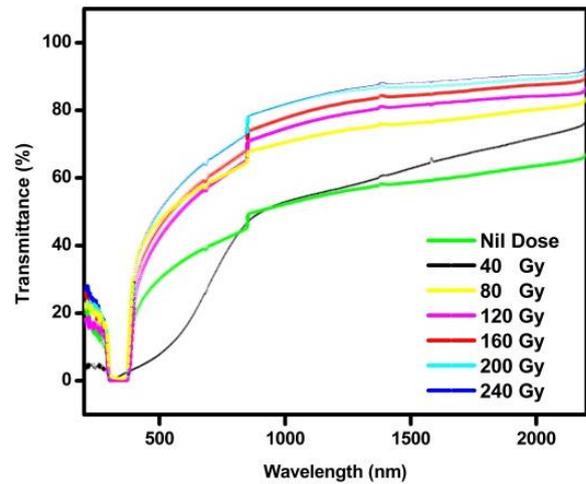


Fig. 4. Transmittance spectra of Pure and gamma irradiated ZnO thin films

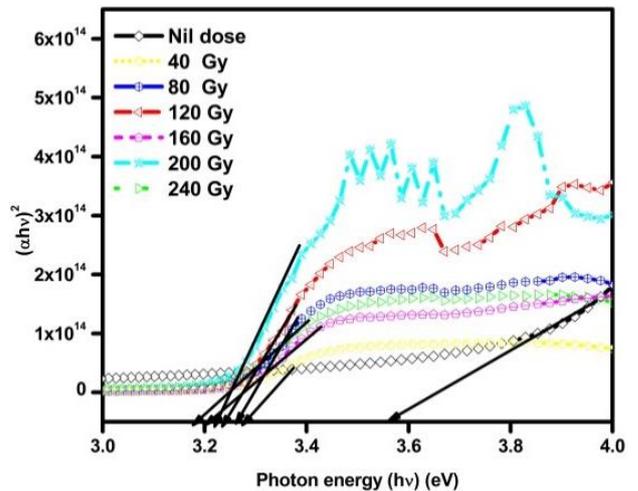


Fig. 5. $h\nu$ Vs $(ah\nu)^2$

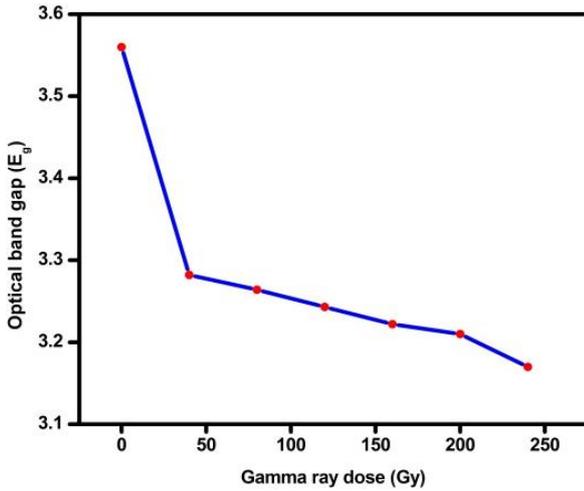


Fig. 6. Gamma ray dose Vs Optical band gap

3.2. X-Ray diffraction studies

The structural elucidation is carried out for un-irradiated and gamma irradiated ZnO thin films. All the films were annealed in air at 450 °C (nil dose) for 15 minutes and then subjected to gamma ray (40 Gy to 240 Gy) and used for the analysis. Fig. 7 & 8 shows the XRD patterns of the pure (nil dose) and gamma irradiated ZnO thin films (240 Gy). The observed diffraction patterns were identified using the JCPDS data [89-1937] and by comparison with earlier reports. The observed d spacing and the respective prominent peaks correspond to the reflections of (1 0 0), (0 0 2), (1 0 1), (1 1 0) and (1 1 2) planes, which coincide well with JCPDS data. Therefore, it has been concluded that the un-irradiated and gamma irradiated films are polycrystalline in nature with hexagonal structure. In the XRD pattern, the presence of the peaks, which correspond to (1 0 0) and (1 0 1) planes confirms the formation of ZnO. A well matching of the observed and standard d values confirms the formation of compound ZnO with hexagonal crystal structure and lattice constant value was found to be 2.8177. The gamma radiation dose increases the peak intensity. Increasing of peak intensity at 240 Gy may be attributed to the increase in the degree of crystallinity. In addition, an average crystalline size (D) of ZnO is estimated from the full width at half maximum (FWHM) of the diffraction peaks at 27.70°, using Scherer equation [16].

$$D = \frac{k\lambda}{\beta \cos \theta}$$

where, the constant k is the shape factor 0.94, λ is the wavelength of X-rays (1.5406 Å for CuK_α), θ is the Bragg's angle and β is the FWHM. The dislocation density (δ) and micro strain (ϵ) can be evaluated from following relations,

$$\delta = \frac{1}{D^2} \text{ lines / m}^2$$

$$\epsilon = \frac{\beta \cos \theta}{4}$$

The crystallite size (D), dislocation density (δ) and micro strain (ϵ) are estimated and presented in Table 1. XRD data shows that a gamma irradiated film is displaying higher intensity than non-irradiated film. The increase in peak intensity for the radiation of 240 Gy is a clear evidence to increased crystallinity and decreased internal micro strain which in turn explains the decrease in the disorder of the films [17].

Table 1. Structural parameters of ZnO thin films

ZnO	(β) X10 ⁻³	(D) nm	(δ) X 10 ¹³	(ϵ) X 10 ⁻³
Nil dose	0.382	21.175	0.00218	0.001622
240 Gy	0.376	21.611	0.00192	0.001602

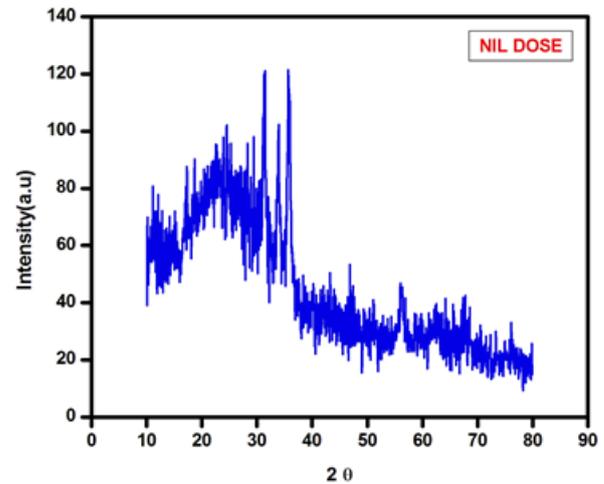


Fig. 7. X-ray pattern for Nil dose

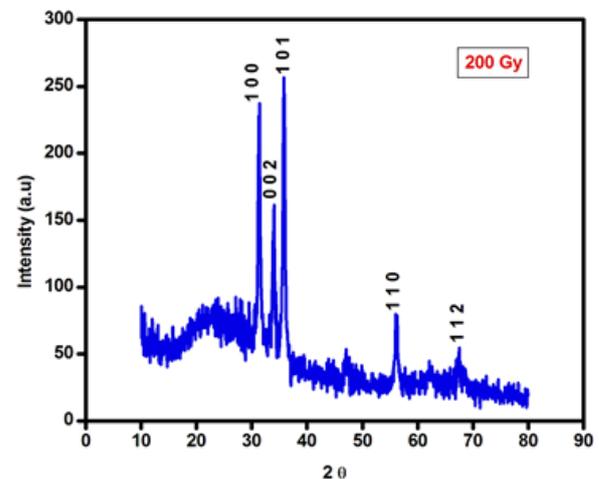


Fig. 8. X-ray pattern for 240 Gy

3.3. Electrical properties

During the exposure of nuclear radiation, dose-rate plays a key role in making excess charge carriers in the film. Again, those charge carriers will recombine within the short period of time once the radiation is switched off. For this reason, the samples were kept at room temperature for 15 min before making any measurements, so that the charge carriers can settle down. DC electrical characteristics of ZnO thin films were measured by Keithley High Resistance Meter / Electrometer 6517B using the two probe arrangement. This measurement was taken at room temperature. This electrometer (6517B) has an in-built capability of output independent voltage source of $\pm 1000\text{V}$. Current was measured for a range of input voltages from 1 to 10 V. I-V measurements for pure and gamma irradiated ZnO thin films are shown in Fig. 9. From the figure, it can be observed that the value of measured current is increasing considerably with an increase in radiation dose up to 240 Gy [4].

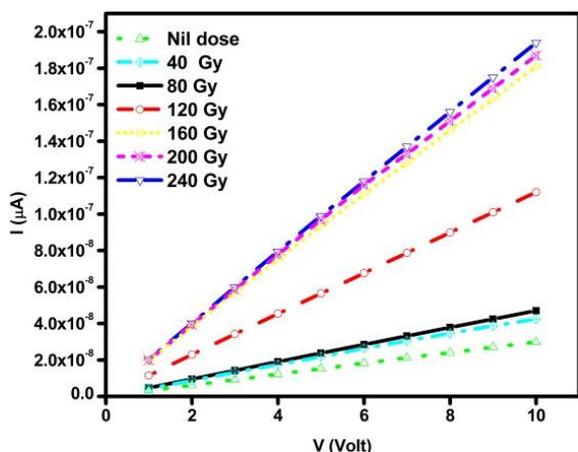


Fig. 9. I-V response of ZnO thin films

4. Conclusion

The X-ray diffraction study shows that the prepared films are in polycrystalline nature with hexagonal structure. The increase in peak intensity and decrease in micro strain was observed for gamma irradiated film owing to the improvement in the crystallinity. The optical studies on spray deposited ZnO thin films exposed to various levels of gamma radiation dose clearly showed that the optical band gap decreases with increase in gamma radiation dose. On the other hand, the current-voltage characteristics show that the current increases almost linearly with the gamma radiation dose up to 240 Gy.

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