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Impact of post-annealing temperature on optical and surface properties of tellurium doped ZnO nanocrystalline films

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Abstract. Investigated in this work is the effect of post-annealing temperature on ZnO nanocrystalline thin films doped with 5 at.% Tellurium. The spin coating method was used to deposit films on the microscopic glass substrate. XRD, AFM, and UV-spectrophotometer were used to characterize the films structure, surface roughness and optical properties. The XRD spectra showed that the nanocrystalline films are of monocrystalline nature. AFM has confirmed the nanocrystalline character of tellurium-doped ZnO. The transmission of exposed films has been decreased with the increase of annealing temperature. The average transmission of all films has been revealed to be higher than 80%. The optical band gap varies slightly with post-annealing temperature.
Keywords: sol-gel, nanocrystallite, annealing temperature, AFM.

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1. Introduction

The *n*-type semiconductor zinc oxide (ZnO) is a promising material for various optoelectronics applications because of its wide band gap (3.3 eV) and outstanding optical and electrical properties at room temperature [1, 2]. The nanocrystalline and particle size of ZnO films play a significant role in increasing the sensitivity of UV sensors that use ZnO as an active material. This is caused by the fact that these films have technological applications, namely: laser diodes, light emitting diodes, solar cells, ultraviolet lasers, and thin film transistors [3, 4]. Pure ZnO nanocrystalline films have various limitations; pure ZnO films have unstable electrical and optical characteristics. As a result, ZnO cannot be applied in its pure form and must be doped with proper chemical elements [5, 6]. For example, the element Tellurium (Te) is regarded as a chalcogen family dopant that can enhance the physical and chemical characteristics of ZnO films. The doped ZnO films are used in a variety of technological applications. Besides, due to their low raw material cost, Te doped ZnO thin films are very interesting [7].

Te doped ZnO nanocrystalline films are obtained using many deposition techniques, such as spray pyrolysis [8], pulsed laser deposition [9], chemical precipitation [10], and sol-gel ones [11] *etc*. Among these methods, sol-gel is particularly useful due to its lower processing temperature, environmental friendliness, and it requires no expensive equipment [12]. It is aimed at making the 5 at.% Te doped ZnO nanocrystalline films produced by varying the post-annealing temperature, which is considered to have a significant role, when using the sol-gel process. Tellurium-doped ZnO films have received little attention yet. Detailed examination of the films was performed to investigate the changes in ZnO structure, surface roughness and optical characteristics with 5% Te doping.

2. Experimental and characterization

2.1. Experimental

Sol-gel spin coating method was used to prepare tellurium doped zinc oxide (ZnOTe) thin films on a microscopic glass substrate. The source materials for Zn and Te are zinc acetate dihydrate ($C_4H_6O_4Zn.2H_2O$) and tellurium dioxide (TeO_2), respectively. Ethanolamine ($H_2NCH_2CH_2OH$) was used as the stabilizer, and 2-methoxyethanol ($C_3H_8O_2$) was used as the solvent. Precursor solutions of 0.4 M for tellurium doped ZnO thin films were prepared by dissolving 5 at.% tellurium tetrachloride powder in zinc acetate dehydrate powder in 2-methoxyethanol and stirring at 80 °C for 30 min with a hot magnetic stirrer. Ethanolamine was then in use as a

stabilizer. The precursor and stabilizer molar ratio was maintained at the level 1:1. Finally, the solution was aged for 12 hours at ambient temperature to produce a clear, homogenous solution. The final solution was applied to clean microscopic glass substrates (1.5×2 cm) using the spin coating technique at 2500 rpm for 25 min. To achieve the desired thickness, the coating technique was repeated ten times. All of the films were preheated at 200 °C for 5 min after each coating. Finally, the post annealing temperature of the spin coated thin films was changed to 225 °C, 275 °C, 325 °C, and 375 °C in a furnace for two hours to explore the influence of the post-annealing temperature on the structural, surface roughness, and optical properties.

3. Results and discussion

3.1. Structural analysis

An X-ray diffractometer (model: Rigaku Miniflex 600) with Cu-K_α radiation (λ = 1.54059 Å) was used to examine the crystalline orientation and the phase structure of the films. Fig. 1 shows the XRD pattern of 5 at.% tellurium doped ZnO nanocrystalline glass film after annealing at 375 °C. The XRD pattern indicates that the 5 at.% Te doped ZnO nanocrystalline film has hexagonal wurtzite structure with matching JCPDS card number 36-1451 [13]. The prominent peak at 34.40° is consistent with hexagonal ZnO in the (002) orientation. For the (002) diffraction peak, the film's full width at half maximum (FWHM) is 0.538°.

The average crystalline grain size (D) was determined using Scherrer's equation from the diffraction peak (002) [13]:

$$D = \frac{\lambda K}{\beta \cos\theta},$$

where λ is the incident X-ray wavelength (1.5405 Å), K = 0.94, β is the Bragg diffraction angle and θ is full width at half maximum (FWHM). The calculated average crystal grain size of the deposited 5% Te doped ZnO nanocrystalline sample was 16.14 nm.

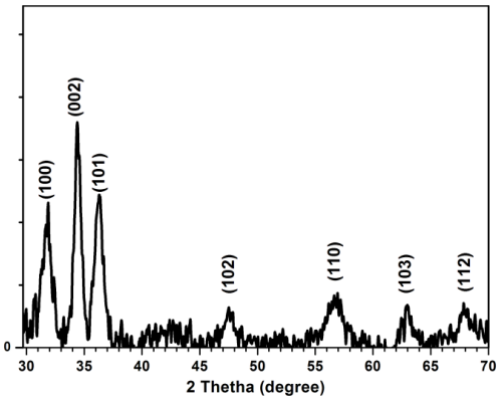


Fig. 1. XRD pattern for 5 at.% Te doped ZnO nanocrystalline film annealed at 375 °C.

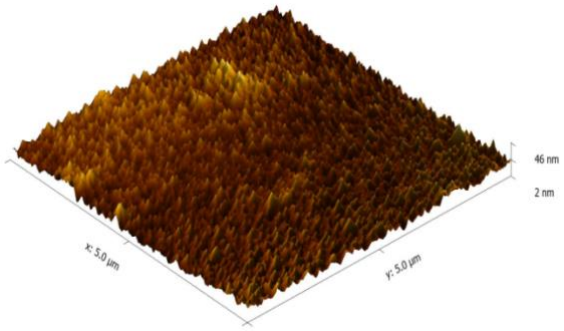
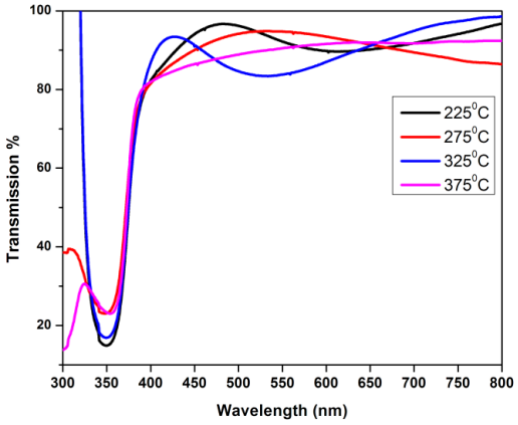


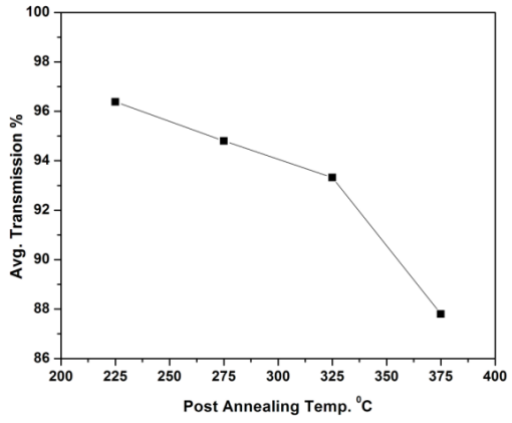
Fig. 2. Surface roughness with the 3D image for 5 at.% Te doped ZnO nanostructure film annealed at 375 °C.

3.2. AFM

The surface morphology of the deposited post-annealed film was investigated using the TriA SPM atomic force microscopy (AFM) instrument. Fig. 2 shows the 3D image of 5 at.% tellurium-doped ZnO nanocrystalline thin film that was post-annealed at 375°C. The scanned area was 50×50 µm, as seen in the image. AFM showed an average surface roughness of 10.8497 nm.



(a)



(b)

Fig. 3. (a) Optical transmittance spectra for the 5% Te doped ZnO nanocrystalline films. (b) A plot of average transmission *vs* post-annealing temperature.

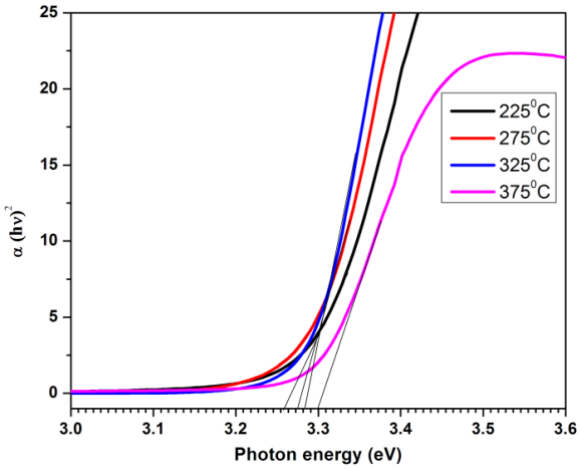


Fig. 4. Tauc's plot for the calculated bandgap energy of 5 at.% Te doped ZnO nanocrystalline films.

3.3. Optical properties

UV–visible spectrophotometer (model: Shimadzu UV-2600) was used to study optical characteristics. Fig. 3a shows the optical transmission spectra from ultraviolet to near-infrared region, which was determined to be ~88% within the range 370 up to 750 nm. The transmittance spectra of Te-doped ZnO nanocrystalline films were examined. Also, the transmittance edges were shifted to lower wavelengths when the annealing temperature of Te doped ZnO nanocrystalline films was increased. Shown in Fig. 3b is a plot of average transmission *vs* post-annealing temperature, the average transparency decreased with increasing the annealing temperature.

The optical bandgap of the 5 at.% Te doped ZnO nanocrystalline films can be calculated using the Tauc equation [14]:

$$(\alpha h\nu)^{1/n} = (h\nu - E_g)\cdot D^{1/n},$$

where $h\nu$ is the photon energy, α is the absorption coefficient, E_g is the optical bandgap, and D is a constant. For a direct transition, the value of n = 1/2 in this equation.

Fig. 4 shows the graph of $(\alpha h\nu)^2$ *vs* photon energy ($h\nu$) of the deposited 5 at.% Te doped ZnO nanocrystalline films. For these films, the bandgaps were calculated and equal to 3.265, 3.300, 3.310 and 3.330 eV. For the Te doped ZnO sample, the bandgap were observed to increase with increasing the annealing temperature and was similar to the results reported by F. Khosravi-Nejad, M. Teimouri *et al.* [15].

4. Conclusion

5 at.% Te doped ZnO nanocrystalline films have been deposited by sol-gel method on glass substrates. Post-annealing temperature treatment has been shown for the structural, surface roughness and optical characteristics of Te doped ZnO films. The XRD spectra imply that the nanocrystalline films are of monocrystalline nature. With increasing the annealing temperature, crystallinity of the Te doped ZnO thin films became improved. The crystalline grain size was found to be close to 16.14 nm. AFM images have shown that the Tellurium doped ZnO films are of nanocrystalline nature. The average transmittance of more than 80% in the visible region was inherent to the deposited films, and their transparency decreased with increasing the post-annealing temperature. The substantial variation in the bandgap value is caused by increasing the post-annealing temperature in the Te doped ZnO structure. Hence, these properties are promising for application in technology of optoelectronic devices.

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Authors' contributions

Anil U. Sonawane: measurements, data analysis, investigation, writing – original draft, writing – review, visualization.

B.K. Sonawane: methodology, validation, data analysis, investigation, resources, data curation, writing – check original draft, writing – editing.

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