International Journal of Science and Engineering and Technology (IJSE)
Maiden Edition
www.federalpolyoko.edu.ng
Volume 1; Issue 2; October 2025; Page No. 96-106.

SYNTHESIS OF ANTIMONY DOPED ZINC OXIDE THIN FILMS USING SOL-GEL DEPOSITION TECHNIQUE

[1]Nwajiobi Chinazo Desmond And [2]Ottih, E.I.
[1]Department of Physics with Electronics, Federal Polytechnic, Oko, Anambra state.
[2]Department of Industrial Physics, Chukwuemeka Odumegwu Ojukwu University, Anambra State.
Corresponding author: *nwajiobichinazo@gmail.com 08060785576*

ABSTRACT
Sol-gel technique was successfully used to deposit antimony zinc oxide (Sb:ZnO) thin films on the pre-treated substrate at 60^{o}C. The deposition time and dopant concentration were varied and its effect on the structural, morphological, compositional and optical properties of the films were examined using X-ray diffraction, Scanning electron microscopy, Energy dispersive X-ray and spectrophotometer respectively. The optical results showed that the deposited films have high absorbance in Ultra Violet region which decrease towards the Near Infra-red region. Energy dispersive X-ray results confirmed that Antimony, Zinc and Oxygen were deposited. XRD result revealed that increase in dip time and dopant concentration led to increase in average crystallite size and improved structure. Dislocation densities and microstrain values decreased with increase in dip time and dopant concentration. X-ray diffraction result revealed hexagon phase of zinc oxide with C-axis orientation. The films have wide energy bandgap which is used for optoelectronic and thermoelectric device fabrication. The grown films have high absorbance in the ultra violet region so are good materials for window coating in the temperate regions of the world, (like in Nigeria) because all the harmful ultra violet rays will be absorbed by such films when used to coat the windows. This is a good substitute of the conventional costly air conditioner.

Keywords: Antimony, zinc oxide, thin films, optical properties, sol-gel technique

INTRODUCTION
Thin film materials play a cruial role in driving ongoing technological progress across optoelectronics, photonics, and magnetic device domains. Research on thin films has notably propelled various areas of solid-state physics and chemistry, owing to phenomena distinctively tied to film thickness, geometry, and structure. The fabrication of materials into thin films facilitates their seamless integration into diverse device types. These films exhibit exceptional thermal stability and reasonable hardness, yet they remain delicate. (Rao and Shekhawat, 2013). Processing materials into thin films simplifies their integration into various devices, and the properties of these materials often exhibit significant variations in thin film form. Functional materials with specific electrical, magnetic, optical properties, or wear resistance are commonly used in thin film applications, leveraging the ability to control properties through film thickness.

Cite this article as:
Nwajiobi, C.D. et al. (2025). Synthesis of Antimony Doped Zinc Oxide Thin Films Using Sol-Gel Deposition Technique. *International Journal of Science and Engineering and Technology (IJSE), 1(2)* 96-106.

Zinc oxide thin film has a wide range of applications in solar cells, optoelectronic devices, gas sensors because of its high transparency and wide energy bandgap. Doping ZnO with a group V element such as Antimony can improve its optical, electrical, structural and compositional properties. Unfortunately getting high quality and uniform antimony doped Zinc oxide thin film with the desired properties is great challenge.

Many deposition methods such as sputtering, chemical vapor deposition, pulsed laser deposition and molecular beam epitaxy have been used to deposit Sb:ZnO but these methods are complex and expensive. Sol-gel deposition technique is a low-cost, low temperature and versaltile method for synthesizing Antimony doped ZnO thin films, but to enhance film crystallinity, optical, electrical and compositional properties there is need to optimize some of the deposition parameters such as doping concentration, chemical environment and annealing temperature. There is need to develop to an optimized sol-gel deposition process to achieve film uniformity, controlled doping and desired optical properties. To resolved the issues relating to film quality, property optimization and film uniformity there is need to examine the synthesis of Sb:ZnO thin films using sol-gel deposition technique. This study aims at investigating the effect of antimony doping concentration and deposition time on the optical, structural and compositional properties Antimony doped zinc oxide thin films using sol-gel deposition method.Many deposition methods such as sputtering, chemical vapor deposition have been recently used to fabricate p-type Zinc oxide films but sol–gel technique has been recognized as one of the most promising techniques to achieve the p-type zinc oxide thin film because of its considerable advantages comparing to other methods including simple deposition setting, ability to design and control chemical composition and maintain a homogeneous composition, formation of uniform mixtures as composite oxides, low-cost system, and low-cost precursor.

Varieties of methods have been used to dope zinc oxide with antimony. Kumar et al (2015) have been able to deposit Antimony (Sb) doped zinc oxide (ZnO) thin films on the glass substrate at 450°C using spray pyrolysis technique. The starting solution of 0.05M concentrations of zinc acetate anhydrous [Zn (CH3COO) 2] in methanol [CH3OH] was used to spray. Antimony acetate [Sb(CH3COO)3] was used as a source of dopant, a little amount of which was added to the starting solution. By changing the weight ratio [Sb/Zn] the doping level was varied. SEM analysis showed a change in surface morphology of Sb doped ZnO thin films. Doping results in a marked increase in conductivity without affecting the transmittance of the films.

Çelik et al. (2020) employed the spin coating technique to fabricate uniform thin films of both pure ZnO and Sb-doped ZnO on soda lime glass substrates at room temperature. They investigated the impact of Sb doping on the structural properties of ZnO thin films. The optical properties of the ZnO thin films with Sb concentrations of 0%, 1%, 2%, and 3% exhibited a significant variation in the optical energy band gap, highlighting the influence of Sb dopants. Furthermore, scanning electron microscopy (SEM) images revealed that pure ZnO thin films exhibited a distinctive nanofiber structure, which gradually disappeared with an increase in the doping ratio, resulting in more homogeneous films.

Cite this article as:
Nwajiobi, C.D. et al. (2025). Synthesis of Antimony Doped Zinc Oxide Thin Films Using Sol-Gel Deposition Technique. *International Journal of Science and Engineering and Technology (IJSE), 1(2)* 96-106.

Sinornate et al (2021) investigated how the annealing atmosphere affects the optical, structural, morphological, and electrical characteristics of Sb-doped ZnO thin films deposited using the Sol-Gel method. X-ray diffraction (XRD) analysis indicated that these films exhibited a pure ZnO hexagonal wurtzite structure without any impurities. The crystallinity of the films deteriorated significantly due to the combined influence of the Sb dopant and the annealing atmosphere, resulting in a reduction in nanoparticle size after annealing in nitrogen and argon atmospheres as well as upon doping. X-ray photoelectron spectroscopy (XPS) results confirmed the successful incorporation of Sb3+ into the ZnO lattice, and the shift in XPS spectra for films annealed in a nitrogen atmosphere suggested the formation of nitrogen bonds with zinc. Photoluminescence (PL) spectra demonstrated a blue shift in the near-band-edge emission, attributed to the substitution of Sb dopant in the Zn site within the ZnO lattice, as well as a red-shifted infrared emission induced by the presence of Sb dopant. Furthermore, Sb doping and annealing in a nitrogen atmosphere led to a decrease in the electrical resistance of the ZnO film.

MATERIALS AND METHODS

The substrates were first subjected to distinct pre-treatment and the beakers washed and distilled with distilled water to ensure no presence of impurities that might increase the film thickness. Antimony doped zinc oxide (Sb:ZnO) sol gel was formed in a 250 mL beaker at 60℃. The reaction mixture contained zinc acetate, antimony trichloride and sodium hydroxide molar solutions. Firstly, 50 mL of 0.2 M of zinc acetate was transferred to the beaker, followed by addition 25 mL of 0.04 M of antimony chloride. The mixture was stirred with a magnetic stirrer for 10 minutes. After that, 50 mL of 0.5 M of NaOH was added drop-wise into the mixture containing zinc acetate and antimony trichloride. The variation in dipping time was achieved by observing dip time intervals of 2 minutes, 4 minutes, 8 minutes, and 10 minutes. Six (6) successive dips were carried to form uniform and adhesive coatings of Sb:ZnO on the surface of the substrates. After the dipping process, the synthesized thin films were annealed at 150 ℃ for 30 minutes. Table 1 shows the constituents of the sol gel bath for the deposition.

Table 1: Variation of dip time for sol gel dip coated Sb:ZnO thin film

Sample	Zn(ace) .2H_2O		$SbCl_3$		NaOH		No. of Dipping	Dip Time (mins)
	mol/dm^3	Vol. (ml)	mol/dm^3	Vol. (ml)	Mol.	Vol. (ml)		
ZB_1	0.20	50.00	0.04	25.00	0.50	50.00	6	2
ZB_2	0.20	50.00	0.04	25.00	0.50	50.00	6	4
ZB_3	0.20	50.00	0.04	25.00	0.50	50.00	6	6
ZB_4	0.20	50.00	0.04	25.00	0.50	50.00	6	8
ZB_5	0.20	50.00	0.04	25.00	0.50	50.00	6	10

To achieve the variation of mole concentration of the dopant, five different concentrations of 0.00M, 0.02 M, 0.04 M, 0.06 M and 0.08 M were used to synthesize antimony doped zinc oxide sol gels. To form adhesive and uniform thin films on the surface of the cleaned microscopic substrate used for the deposition, six (6) successive dips were carried out from each of the synthesized sol gel bath. The dips were carried out at intervals of 2 minutes. After the dipping process, the synthesized thin films were annealed at 150 ℃ for 30 minutes. Table 2 shows the constituents of the sol gel bath for the deposition.

Cite this article as:
Nwajiobi, C.D. et al. (2025). Synthesis of Antimony Doped Zinc Oxide Thin Films Using Sol-Gel Deposition Technique. *International Journal of Science and Engineering and Technology (IJSE), 1(2)* 96-106.

Table 2: Variation of dopant concentration for sol gel dip coated Sb:ZnO thin film

Sample	Zn(ace) .2H_2O		$SbCl_3$		NaOH		No. of Dipping	Dip Time (mins)
	mol/dm^3	Vol. (ml)	mol/dm^3	Vol. (ml)	Mol.	Vol. (ml)		
ZB_6	0.20	50.00	-	-	0.50	50.00	6	2
ZB_7	0.20	50.00	0.02	25.00	0.50	50.00	6	2
ZB_8	0.20	50.00	0.04	25.00	0.50	50.00	6	2
ZB_9	0.20	50.00	0.06	25.00	0.50	50.00	6	2
ZB_{10}	0.20	50.00	0.08	25.00	0.50	50.00	6	2

The amount of salt used was calculated using equation (1) given below

$$R_{mass} = \frac{\text{Molarity x Molar Mass x Volume}}{1000\text{cm}^3} \quad 1$$

The deposited films' thicknesses (t) were evaluated using the gravimetric method given by

$$t = \frac{\Delta m}{\rho A}, \quad 2$$

where Δm is the mass of the film; A is the surface area of the deposited film and ρ is the bulk density of the material film.

The optical absorbance values of these sol gel deposited thin films were obtained using spectrophotometer (model: 756S UV – VIS) at Nano Research Laboratory, Department of Physics and Astronomy, University of Nigeria Nsukka, Enugu State, Nigeria. Other optical properties of the films such as transmittance, reflectance, refractive index, extinction coefficient and energy band gap were calculated as follows.

Transmittance, $T = 10^{-A}$ 3

Reflectance, $R = 1 - (A + T)$ 4

Refractive index, $\eta = \frac{(1+\sqrt{R})}{(1-\sqrt{R})}$ 5

Extinction coefficient, $\mathrm{k} = \frac{\alpha\lambda}{4\pi}$ 6

Energy band gap, $(\alpha hv)^n = \beta\left(hv - E_g\right)$. 7

Crystal structural analyses of the deposited thin films were done using X– ray diffractometer machine. The crystallite sizes and microstrains of the films were evaluated using Scherrer's formula. Debye – Scherrer's formula for calculating crystallite sizes of a thin film material is given by (Ravindranah et al, 2016; Okoriemoh, et al., 2019) as

Cite this article as:
Nwajiobi, C.D. et al. (2025). Synthesis of Antimony Doped Zinc Oxide Thin Films Using Sol-Gel Deposition Technique. *International Journal of Science and Engineering and Technology (IJSE), 1(2)* 96-106.

$$D=(0.9\ \lambda)/\beta\cos\theta. \quad 8$$

The dislocation density (δ) of thin films can be estimated using expression as provided by Anbarasi et al., (2016); Hadri et al., (2015) in equation (9).

$$\delta = \frac{1}{D^2}. \quad 9$$

Micro-strain (ε) of the thin film sample can be estimated using the expression in equation (10) as given by (Awada et al., 2020 and Hadri et al., 2015).

$$\varepsilon = \frac{\beta}{4tan\theta} \quad 10$$

RESULTS AND DISCUSSIONS

A. Optical properties

The optical absorbance values of these sol gel deposited thin films were obtained using spectrophotometer (model: 756S UV – VIS). Figure 1 shows the variation of the thin film thickness with deposition time (fig 1a) and dopant concentration (fig 1b). It was recorded that the film thickness increases as the deposition time and dopant concentration increase. Figure 2 shows the optical absorbance of the Sb:ZnO films and it was observed that the deposited films have high absorbance in the ultra violet region and it can also be seen that the absorbance increases with increase in deposition time and dopant concentration and decreases with increase in wavelength. It is observed from figure 3 that transmittance decreased with increase in deposition time and dopant concentration but increased with increase in wavelength. The reflectance is maximum in UV region and minimum within NIR region. The dopant concentration of 0.08M recorded the maximum reflectance value of 20.34% in the UV region while the dopant concentration of 0.00M recorded the minimum reflectance value of 7.99% within NIR region (figure 4). It can be seen from figure 5 that the bandgap decreases as the dip time increases.

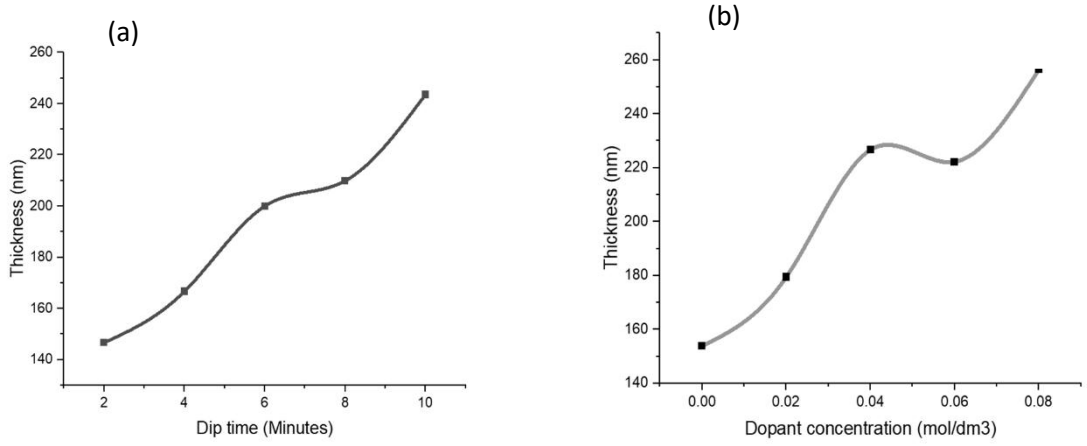


Figure 1: **Graph of film thickness versus dip time (1a) and dopant concentration (1b).**

Cite this article as:
Nwajiobi, C.D. et al. (2025). Synthesis of Antimony Doped Zinc Oxide Thin Films Using Sol-Gel Deposition Technique. *International Journal of Science and Engineering and Technology (IJSE), 1(2)* 96-106.

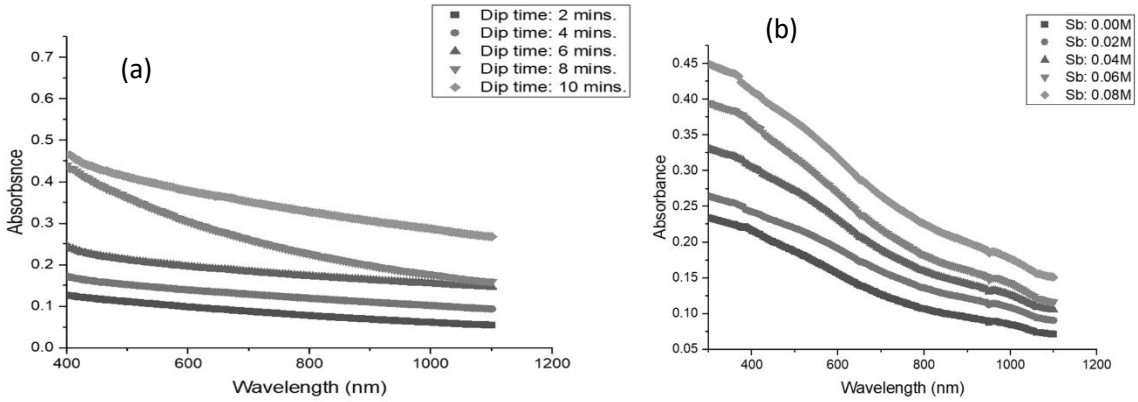


Figure 2: **Graph of absorbance against wavelength for Sb:ZnO thin films with variation in deposition time (2a) and dopant concentration (2b).**

There are great improvements in the optical properties of the deposited film with the incorporation antimony starting from absorbance, transmittance, reflectance etc.

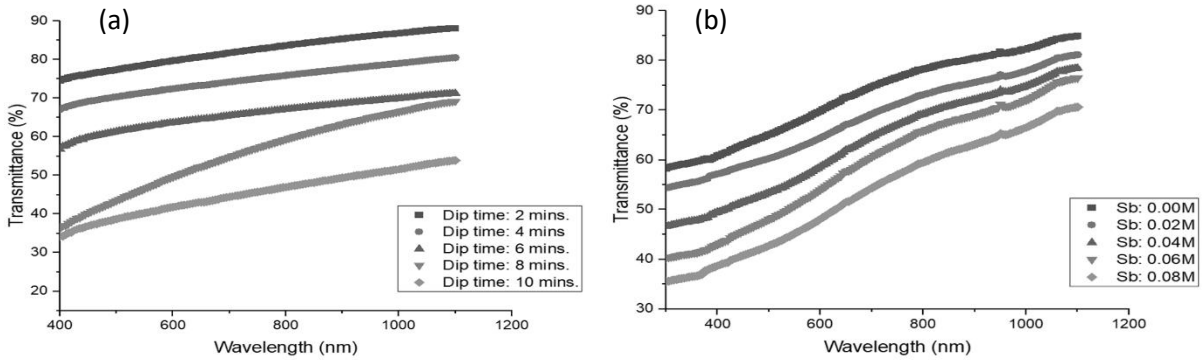


Figure 3: **Graph of transmittance against wavelength for Sb:ZnO thin films with variation in deposition time (3b) and dopant concentration (3b).**

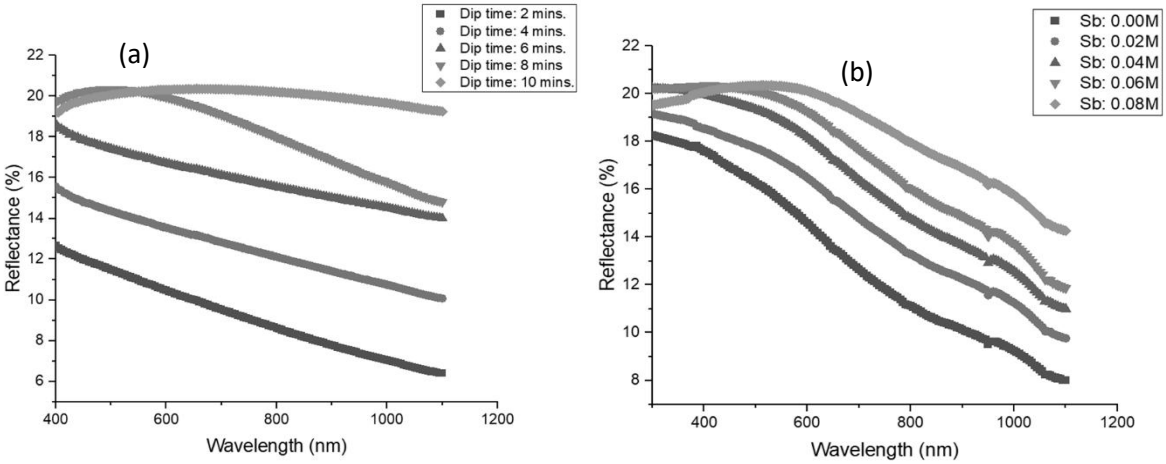


Fig 4: **Graph of reflectance against wavelength for Sb:ZnO thin films with variation in deposition time (4a) and dopant concentration (4b).**

Cite this article as:
Nwajiobi, C.D. et al. (2025). Synthesis of Antimony Doped Zinc Oxide Thin Films Using Sol-Gel Deposition Technique. *International Journal of Science and Engineering and Technology (IJSE), 1(2)* 96-106.

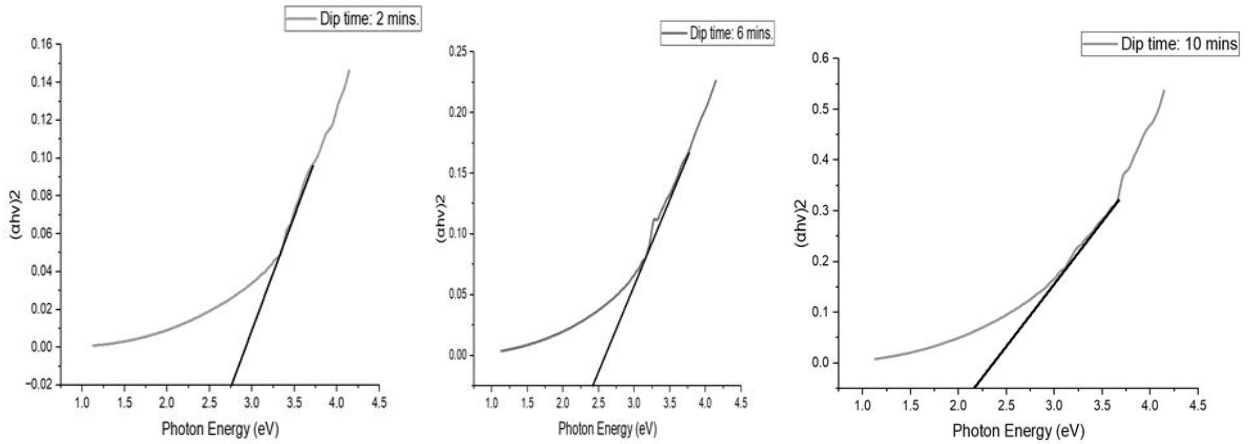


Fig 5: **Graphs of $(\alpha hv)^2$ against photon energy (eV) of Sb:ZnO thin film for different dip time.**

B. Structural Properties

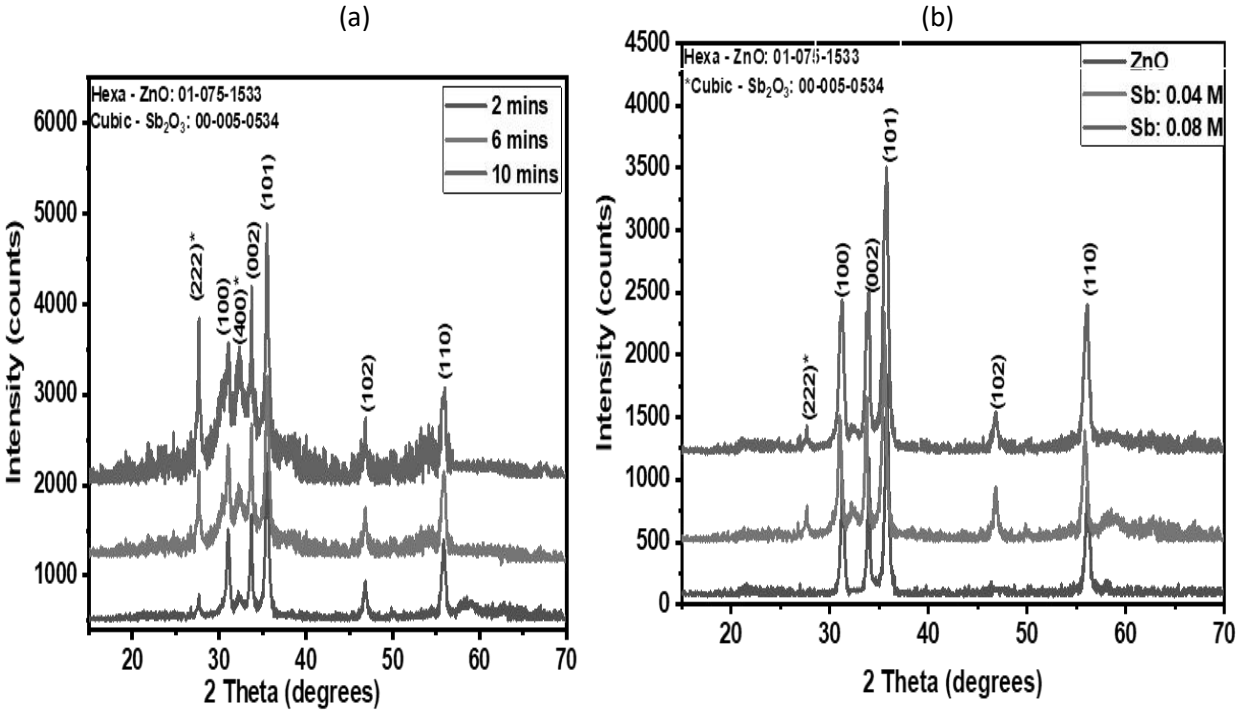


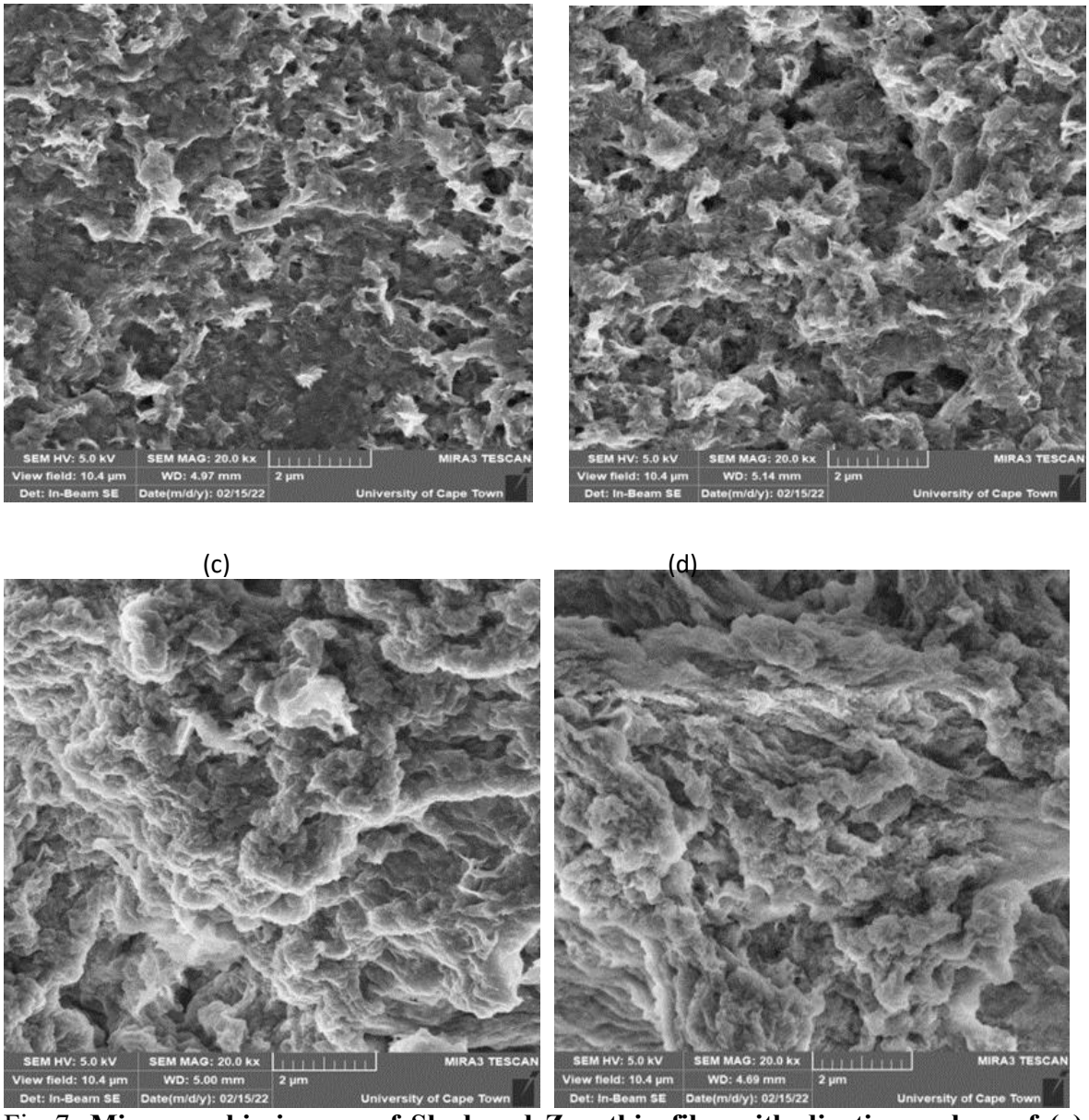
Fig 6: X – ray diffraction pattern of antimony doped zinc oxide (Sb:ZnO) thin films formed at different dip time of 2 minutes, 6 minutes and 10 minutes (5a) and (Sb:ZnO) thin films deposited with different concentration of antimony precursor (5b).

The XRD pattern shows that the deposited Sb:ZnO thin films contained mixed structural phases of hexagonal ZnO and cubic Sb2O3 corresponding to JCPDS file number (01-075-1533) and (00-005-0534). The intensities of the x – ray diffraction spectra were found to increase as dip time increased from 2 minutes to 10 minutes. The average crystallite sizes of 18.780 nm, 21.546 nm, and 23.158 nm were obtained for Sb:ZnO thin films deposited under 2 minutes, 6 minutes and 10 minutes respectively (figure 6a). From 6b the pattern shows that the films deposited at different deposition time have diffraction peaks at 31.289 °, 33.947 °, 35.777 ° and 56.124 °

Cite this article as:
Nwajiobi, C.D. et al. (2025). Synthesis of Antimony Doped Zinc Oxide Thin Films Using Sol-Gel Deposition Technique. *International Journal of Science and Engineering and Technology (IJSE), 1(2)* 96-106.

which correspond to miller indices of (100), (002), (101) and (110). The structural pattern observed in the deposited ZnO thin film is attributed to hexagonal phase of zinc oxide with JCPDS file number (01-075-1533). The results show that crystallite sizes of the films increase as the deposition time andantimony precursor increase. Also, dislocation densities and microstrain values decrease as deposition time and antimony precursor increase.

(a) (b)



(c) (d)

Fig 7: **Micrographic images of Sb doped Zno thin film with dip time values of (a) 6 minutes (b) 10 minutes and dopant concentration (c) 0.04M (d) 0.08M**

Figure 7 shows micrographic images of Sb:ZnO deposited on a glass substrate with different deposition time (fig 7a & 7b) and dopant concentration (fig 7c &7d). The image reveals a granular microstructure and a network of interconnected pores and ridges.

C. Compositional properties

Cite this article as:
Nwajiobi, C.D. et al. (2025). Synthesis of Antimony Doped Zinc Oxide Thin Films Using Sol-Gel Deposition Technique. *International Journal of Science and Engineering and Technology (IJSE), 1(2)* 96-106.

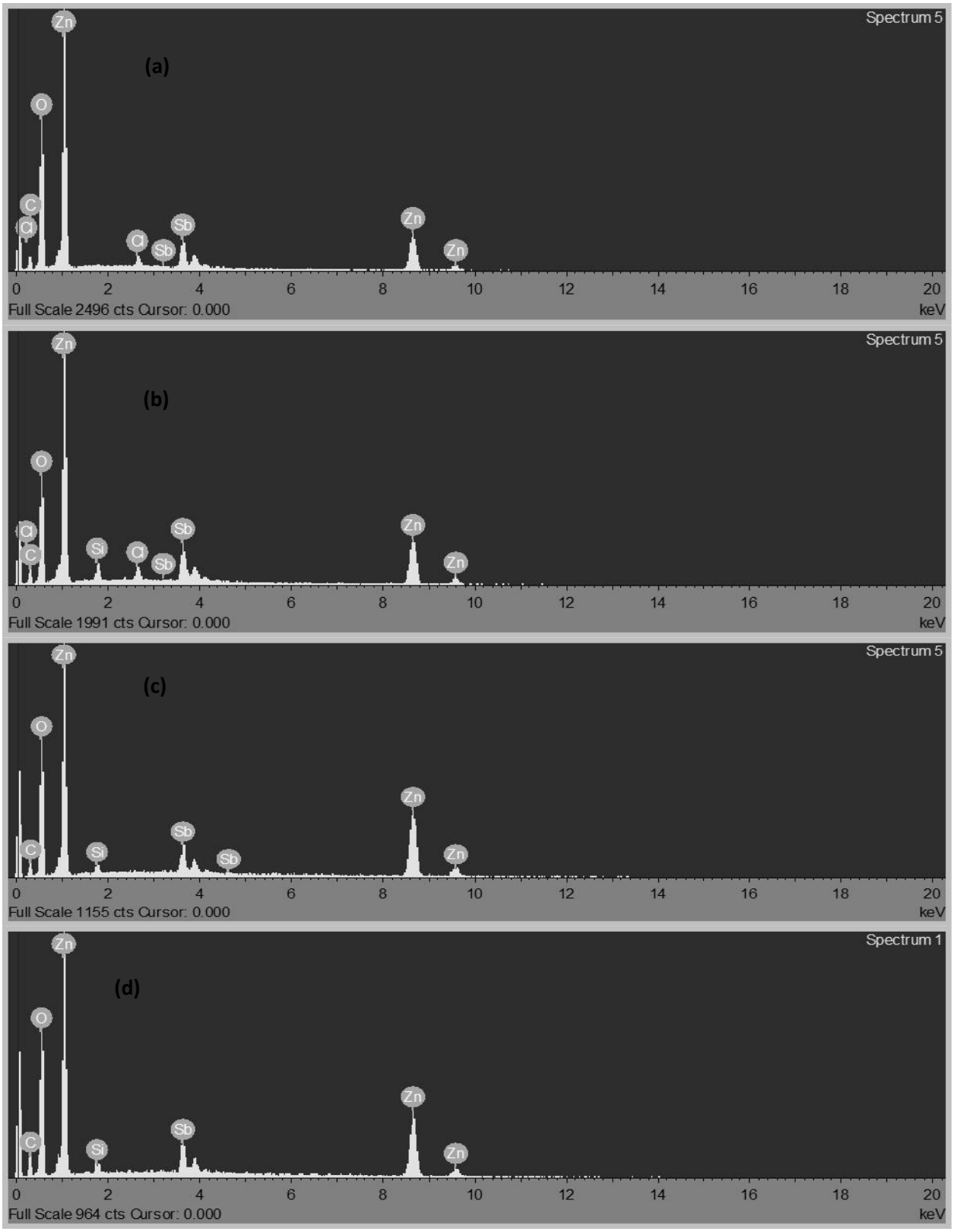


Fig 8: **Compositional structures of Sb:ZnO thin film for dipping time of (a) 6 minutes (b) 10minutes and dopant conc. of (c) 0.04M (d)0.08M**

Cite this article as:
Nwajiobi, C.D. et al. (2025). Synthesis of Antimony Doped Zinc Oxide Thin Films Using Sol-Gel Deposition Technique. *International Journal of Science and Engineering and Technology (IJSE), 1(2)* 96-106.

Figure 8 shows the compositional structures of Sb:ZnO thin films with different dip times of (a) 2 minutes, (b) 6 minutes, (c) 10 minutes (fig 8a & 8b) and with different dopant concentration of (a) 0.00M, (b) 0.04M, (c) 0.08M. These deposited films on the substrate revealed the presence of Oxygen, Zinc, Antimony and some impurities such as Carbon, Magnesium, Silicon, Chlorine and calcium. It was observed that the longer the dip time the more amount of Zinc and Antimony atoms deposited on the substrate and same happens as the dopant concentration increases.

Table 3: Compositional analysis of Sb:ZnO thin film for dopant conc. of (a)0.00M (b) 0.04M (c)0.08M

Sample	Sb^{3+}Conc	Spectrum	C	O	Si	Cl	Zn	Sb	Total
ZB 6	0.00	Spectrum1	11.62	33.27	0.37	1.77	42.24	10.73	100
ZB 8	0.04	Spectrum5	7.66	31.58	0.88	Nil	51.25	8.63	100
ZB 10	0.08	Spectrum1	12.76	33.65	0.84	Nil	43.38	9.38	100

Table 4: Compositional analysis of Sb:ZnO thin film for dipping time of (a) 2 minutes (b) 6 minutes (c) 10minutes.

Sample	Dip time (mins)	Spectrum	C	O	Mg	Si	Cl	Ca	Zn	Sb	Total
ZB1	2	Spectrum5	12.75	58.75	1.08	12.97	0.97	1.72	7.34	4.43	100
ZB3	6	Spectrum5	10.49	41.25	Nil	Nil	1.31	Nil	35.33	11.62	100
ZB5	10	Spectrum5	11.57	31.47	Nil	2.04	1.79	Nil	41.41	11.72	100

CONCLUSION AND RECOMMENDATION

The effect of the deposition time and dopant concentration on the optical, structural and compositional properties of Sb:ZnO has been investigated. The deposited films have a hexagonal phase of zinc oxide same as Sb:ZnO films prepared by Kumar et al., (2015). Using spray pyrolysis techniques. Sb:Zno prepared by Junrez et al., (2014) also showed same pattern and c-axis orientation as films grown in this work. The XRD pattern of Sb:ZnO thin films deposited with different concentration of Antimony precursor show that the deposited ZnO film has diffraction peaks at 30.289o, 33.947o; 35.777o and 56.124o which correspond to miller indices of (100), (002), (101), and (110). This is almost similar to the one obtained from the XRD pattern of Sb:ZnO prepared by Sinornate et al., (2021). The grown films have high absorbance in the ultra violet region so are good materials for window coating in the temperate regions of the world, (like in Nigeria) because all the harmful ultra violet rays will be absorbed by such films when used to coat the windows. This is a good substitute of the conventional costly air conditioner. Hence doping Zinc oxide thin film with Antimony improves its optical properties as can be seen from the results of this study. It has also been shown from this study that sol-gel deposition technique is an effective deposition method to improve the quality and optical properties of deposited films by varying their deposition time and dopant concentration.

Cite this article as:
Nwajiobi, C.D. et al. (2025). Synthesis of Antimony Doped Zinc Oxide Thin Films Using Sol-Gel Deposition Technique. *International Journal of Science and Engineering and Technology (IJSE), 1(2)* 96-106.

It is recommended that other thin film properties like electrical and magnetic properties should be investigated to ascertain their possible application. Also other growth parameters apart from deposition time and dopant concentration should be optimized to know their effect on the formation of the films and optical, structural, morphological and compositional properties.

REFERENCE

ÇelikÖmer ,ŞilanBaturay & Yusuf SelimOcak.(2020). Sb doping influence on structural properties of ZnO thin films, Materials Research Express 7 026403.received 24 October 2019 revised 9 January 2020 accepted for publication 16 January 2020 published 4 February 2020. https://doi.org/10.1088/2053-1591/ab6c8a

Anbarasi, M., Nagarethinam, V. S., Baskaran, R. and Narasimman, V. (2016). Studies on the structural, morphological and optoelectrical properties of spray deposited CdS:Pb thin films. *Pacific Science Review A: Natural Science and Engineering*, 18(1), 72 – 77.

Awada, C., Whyte, G. M., Offor, P. O., Whyte, F. U., Kanoun, M. B., Goumri-Said, S., Alshoaibi, A., Ekwealor, A. B. C., Maaza, M. & Ezema, F. I. (2020). Synthesis and Studies of Electrodeposited Yttrium Arsenic Selenide Nanofilms for Opto-Electonic Applications. *Nanomaterials*, 10(8), 1557.

Hadri, A., Nassiri, C., Chafi, F. Z., Loghmarti, M. and Mzerd, A. (2015).Effect of Acetic Acid Adding on Structural, Optical and Electrical Properties of Sprayed ZnO Thin Films. *Energy and Environment Focus*, 4(1), 12 – 17.

Juarez, G. Diaz, J. Martines-Juarez, P. Lopez Salazar, R. Pena-Sierra, J.I Contreras-Roscon, J. Diaz-Reyes, S. (2014). Structural and Electrical Characterization of Zinc Oxide Doped with Antimony. Mex. Sci. Eng. 1(2014). 35-39.

Kumar Sadananda. N, Kasturi V. Bangera, and G. K. Shivakumar,(2014).Properties of Antimony Doped ZnO Thin Films Deposited by SprayPyrolysis Technique. spectroscopy, interaction with radiation. Received September 18, 2013; accepted for publication December 2, 2014, ISSN 10637826, Semiconductors, 2015, Vol. 49, No. 7, pp. 899–904. © Pleiades Publishing, Ltd., 2015. DOI: 10.1134/S1063782615070143

Okorieimoh, C.C., Chime, U., Nkele, A. C., Nwanya, A. C., Madiba, I. G., Bashir, A. K. H., Botha, S., Asogwa, P. U., Maaza, M., and Ezema, F. I. (2019). Room-temperature synthesis and optical properties of nanostructured Ba-Doped ZnO thin films. *Superlattices and Microstructures*, 130, 321 – 331.

Sinornate Wuttichai, Hidenori Mimura, and Wisanu Pecharapa,(2021).Structural, Morphological, Optical, and Electrical Properties of Sol–Gel Derived Sb-Doped ZnO Thin Films Annealed Under Different Atmospheres. Phys. Status Solidi A 2021, 218, 2000233. https://doi.org/10.1002/pssa.202000233.