



# SYNTHESIS AND CHARACTERIZATION OF OPTICAL AND STRUCTURAL PROPERTIES OF INORGANIC AND GREEN LEAF DOPED SnO THIN FILMS DEPOSITED USING SPRAY PYROLYSIS

C.N. UKWU, J E. EKPE, A.N. NWACHUKWU, E.P. OBOT, O.C. OKAFOR, T.O. DANIEL  
J.C. UMUNNA, P.B. OTAH, B.J. ROBERT, C.A. ELEKWA AND B.J. IFEANYICHUKWU

Email: [ukwunweke@yahoo.com](mailto:ukwunweke@yahoo.com)

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## ABSTRACTS

Dye-sensitized Nanohybrid  $Zn_xSn_yO_z$  thin films were deposited on glass substrates using spray pyrolysis method. The dye extract from the leaves of *tectona grandis* was used. A concentration of 0.1M of  $Zn^{2+}$  and 1% was used respectively. Depositions were carried out at different substrate temperature of 50°C, 100°C, and 150°C. The effect  $Zn^{2+}$  ion and dye extract from *tectona grandis* leaves on optical and solid state properties of the films were examined and analysed. The result showed that the absorbance of the undoped SnO thin films at various substrate temperatures vary from about 0.1-0.7. The absorbance generally increased with deposition/ substrate temperature exhibiting a maximum for films deposited at 150°C. The average transmittance of both un-doped and  $Zn^{2+}$  doped SnO thin films at 350nm is above 90% regardless of the film thickness. The dye doped samples showed an improvement in optical transmission at 625nm. The reflectance spectra of all films exhibited a similar trend. Peak reflectance was observed at 350nm for un-doped and  $Zn^{2+}$  doped samples of SnO thin films while peak reflectance can be observed at 625nm for dye doped samples. It is also observed that the band gaps of the dye doped samples are lower: 1.55eV- 1.83eV than those of the  $Zn^{2+}$  doped samples: 1.60eV – 2.20eV. This showed that the incorporation of the dye shifted the fundamental absorption edge of the un-doped SnO thin films thus providing tuning effect of the band gap for device applications. The diffractograms of SnO doped Zn are characterized by relatively low intensity for all the samples irrespective of the doping concentration compared to the un-doped SnO samples. Obviously, the doping of SnO with 1% dye extracts modified the XRD patterns of the un-doped SnO samples. Substrate temperature also affected the structural properties of SnO:dye sample depicting increase in intensity vis-à-vis crystallinity of the films with increase in substrate temperature. The results equally indicate that there was an increase in the grain sizes that resulted in a decrease in energy badgap of the samples.

**KEYWORDS:** Spray pyrolysis, Optical and Structural (XRD) properties, Dopants and Temperature

## INTRODUCTION:

Organic photovoltaic (OPV) cells or dye-sensitized solar cells (DSSC) are the third generation of solar cell technology. Dye-sensitized solar cell has a number of attractive features. It provides flexible solar modules.

It can be fabricated using conventional roll-printing techniques that are less expensive than the conventional method. The novel in solar cell technology is that portable electronic devices could be re-charged using a low light level potentials and partial shading like solar panels [6].

**C.N. Ukwu**, Alex Ekwueme Federal University Ndufu Alike, P.M.B. 1010, Abakaliki, Ebonyi State

**J E. Ekpe**, Alex Ekwueme Federal University Ndufu Alike, P.M.B. 1010, Abakaliki, Ebonyi State

**A.N. Nwachukwu**, Alex Ekwueme Federal University Ndufu Alike, P.M.B. 1010, Abakaliki, Ebonyi State

**E.P. Obot**, Alex Ekwueme Federal University Ndufu Alike, P.M.B. 1010, Abakaliki, Ebonyi State

**O.C. Okafor**, Alex Ekwueme Federal University Ndufu Alike, P.M.B. 1010, Abakaliki, Ebonyi State

**T.O. Daniel**, Alex Ekwueme Federal University Ndufu Alike, P.M.B. 1010, Abakaliki, Ebonyi State

**J.C. Umunna**, Alex Ekwueme Federal University Ndufu Alike, P.M.B. 1010, Abakaliki, Ebonyi State

**P.B. Otah**, Industrial Physics, David Umahi Federal University of Health Science

**B.J. Robert**, Akanu Ibiam Federal Polytechnic, Unwana

**C.A. Elekwa**,

**B.J. Ifeanyichukwu**,

Dye-sensitized cells are very strong absorbents of light as they harness larger amounts of sunlight more than silicon based solar cells. [10] Hence, the major problem of this study is to investigate the effect of Zinc ion ( $Zn^{2+}$ ) and natural dye extract from fresh leaves of *tectona grandis* (teak plant) on Tin oxide (SnO) as transparent conductive oxide (TCO). Transparent conductive oxide (TCO) films are widely used in many fields including optoelectronic devices. Indium tin oxide is the best transparent conductive oxide (TCO) films that are known for its excellent optical and electrical properties. However, Indium used in indium tin oxide is a scarce resources and expensive. [16] and [8]. Doping with selective elements help to enhance and control the structural, electrical and optical properties of SnO thin films. In addition, doping improves the number of charge carriers and the conductivity of SnO films, for potential application in optoelectronic devices especially in solar cells. Hence, alternative dopants were sorted using  $Zn^{2+}$  and dye extract from the leaves of *tectona grandis* dye extract on SnO thin films. Tin oxide (SnO) and tin-doped indium oxide (ITO) layers, are widely used as transparent conductive oxide (TCO) layers in solar cells due to their superior electrical and optical properties. Generally, SnO is a wide band gap semiconductor with an energy gap of 2.5eV and 3eV. [13], [4].

In this study SnO, doping is achieved by adding  $Zn^{2+}$  atom and dye extract from the leaves of *tectona grandis* into the compound using simple, vacuum free and cost effective spray pyrolysis method in order to tune the optical properties of SnO. The optical properties of the grown layers were examined and analysed. The significance of this study includes: It highlights the effects of dye extract of *tectona grandis* on thin film deposition. It equally x-rays the effect of impurities and temperature on tin oxide thin film properties. This study uncovers the pivotal role of impurity and temperature in ensuring better utilization of material in producing cheaper and more efficient thin films of high qualities.

#### Experimental details and characterization

Aqueous solution of 0.1M of Tin (II) chloride ( $SnCl_2$ ) was prepared by dissolving 19gram of the salt in 100ml of distilled water as a source of SnO. Aqueous solution of 0.1M Zinc Chloride was prepared by dissolving 1.36g in 100ml of distilled water as a source

of  $Zn^{2+}$  (dopants), Sodium hydroxide was used as source of  $OH^-$  and dye extract of *Tectona grandis* as dopant. Microscopic glass slide of dimension 26mm X 76mm, thickness – 1.0mm and refractive index – 1.52 was used as the substrate. The substrates were cleaned with acetone and cotton to remove visible contamination such as dust. Then the substrates were immersed in ethanol solution for 5 hours followed by washing with distilled water in order to remove surface oxides and precipitation. The substrates were finally ultrasonically cleansed and oven dried.

To obtain SnO thin films 40ml of 0.1M aqueous solution of Tin (II) chloride and 4 drops of aqueous ammonia was measured into 50ml glass beaker. The mixture was stirred under highspeed technique for about 5minutes using magnetic stirrer in order to obtain uniform solution. Under the stirring 10ml of the solution was measured out using pump syringe into spray pyrolysis sample bottle which was fastened on valve rod of air brush and sprayed for 60 seconds on heated substrate at the temperature of 50°C using the compressor with the air brush at pressure of 22 Pa. These were repeated for 2 different 10ml of the solution at 100°C and 150°C respectively at deposition angle of 28°, height of 36m, distance -----and time of 60seconds.

The dopants or impurities are normally 10% of chosen volume of the solution in conventional chemical experiment. To obtain SnO:Zn, 1ml of 0.1M of  $ZnCl_2$  was mixed with 18ml of 0.1M of  $SnCl_2$  and to the mixture was added 1ml of NaOH. Similarly, to obtain SnO:dye, 1 % of the dye was added to a mixture of 1ml of 0.1M of  $ZnCl_2$  and 18ml of 0.1M of  $SnCl_2$  The mixture was stirred using a magnetic stirrer for 30 minutes in order to obtain homogenous solution. After the stirring, 10ml of the resulting mixture was measured into sample bottle that was fixed on nozzle valve and sprayed on heated substrate temperatures of 50°C, 100°C and 150°C.

Optical properties of the films were measured using Thermo scientific GENESYS 10S model UV-VIS spectrophotometer. Other optical and solid state parameters were deduced from the transmittance and reflectance spectra.

#### RESULTS AND DISCUSSION

The films grown were milky white in appearance which gradually turned to brown colour with the increase of temperature. All the deposited films are pore free, uniform and strongly adherent to the substrate surface.

Figures 1 to 3 shows transmittance Vs wavelength for SnO, SnO:Zn, and SnO:Dye-extract

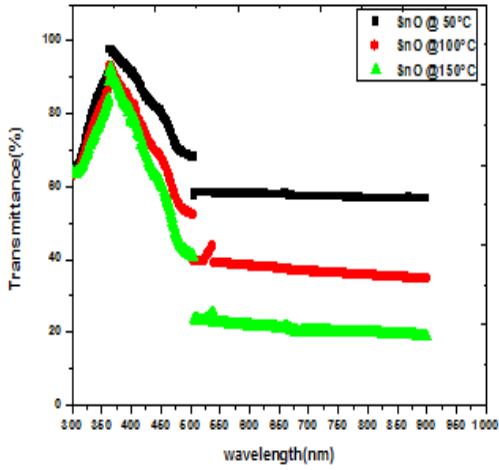


Fig.1. Transmittance Vs Wavelength for SnO (undoped)

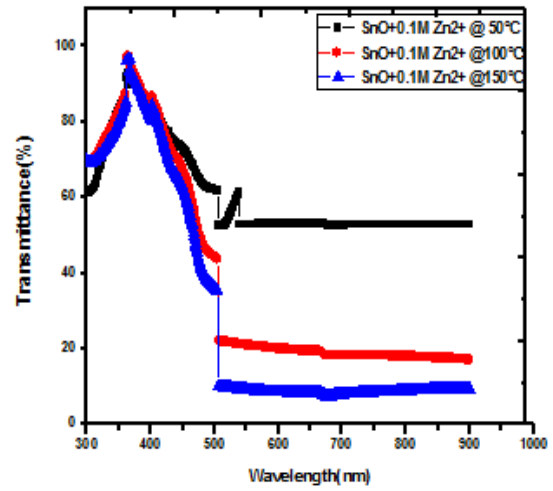


Fig.2. Transmittance Vs Wavelength for SnO+0.1M of Zn<sup>2+</sup> (doped)

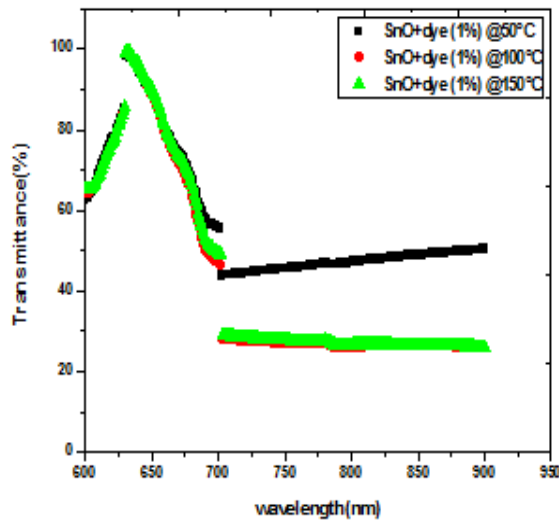


Fig. 3. Transmittance Vs Wavelength for SnO + dye (1%) (doped)

Figures 4 to 6 shows Absorbance Vs Wavelength for SnO, SnO:Zn, and SnO:Dye-extract

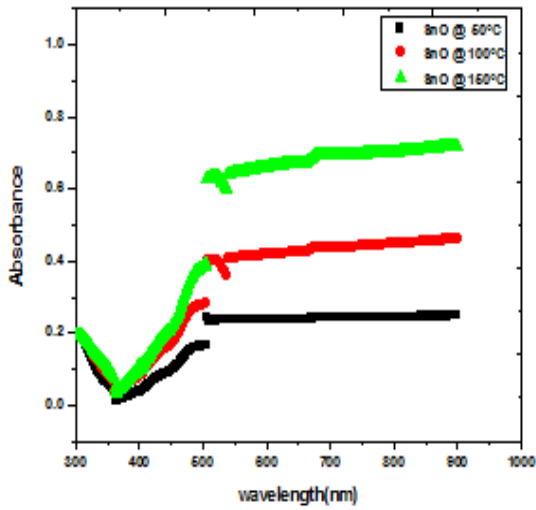


Fig.4. Absorbance Vs Wavelength for SnO (undoped)

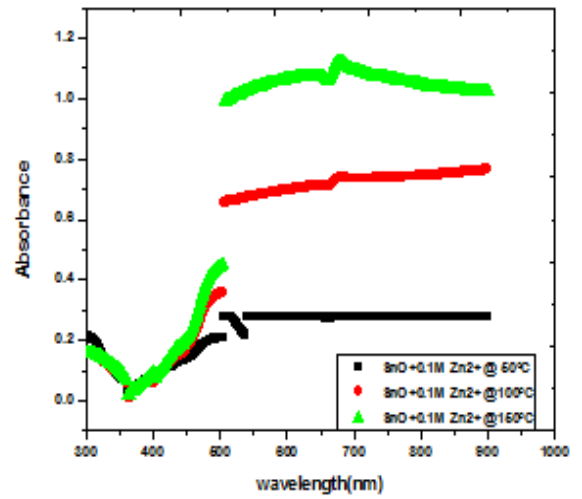


Fig.5. Absorbance Vs Wavelength for SnO+0.1M of Zn<sup>2+</sup> (doped)

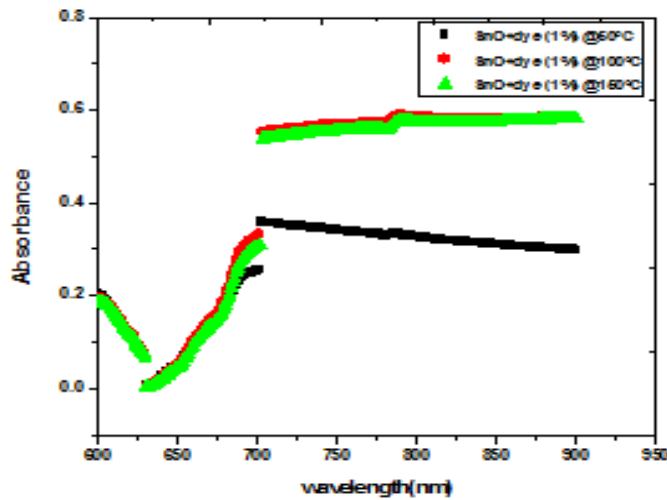


Fig.6. Absorbance Vs Wavelength for SnO+ 1% dye (doped)

Figures 7 to 9 shows Reflectance Vs Wavelength for SnO, SnO:Zn, and SnO:Dye-extract

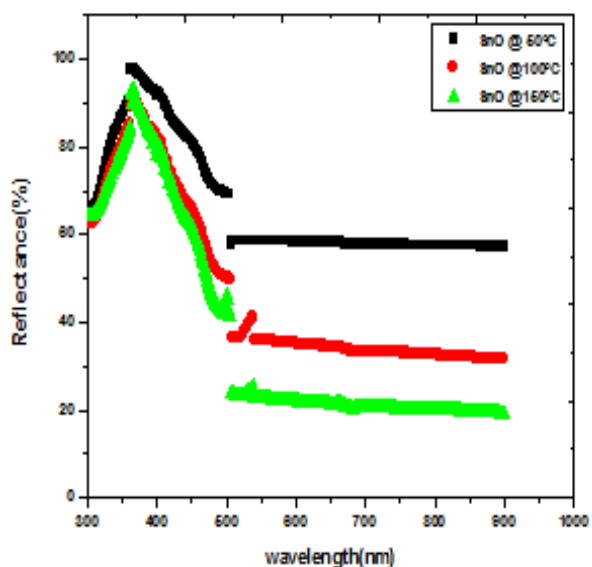


Fig. 7. Reflectance (%) Vs Wavelength for SnO (undoped)

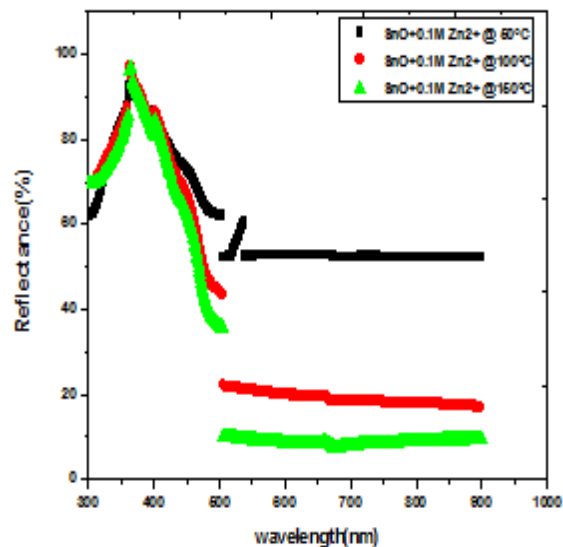


Fig.8. Reflectance (%) Vs Wavelength for SnO+ 0.1of Zn<sup>2+</sup> (doped)

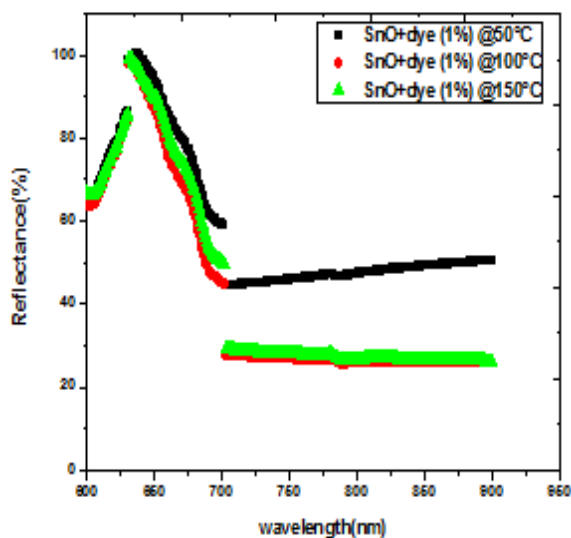


Fig. 9. Reflectance Vs Wavelength for SnO+ dye (1%) dye (doped)

Figures 10 to 12 shows  $(\alpha h\nu)^2$  ( $\text{eV}^2\text{m}^{-2}$ ) Vs  $h\nu$ (eV) for SnO, SnO:Zn, and SnO:Dye-extract

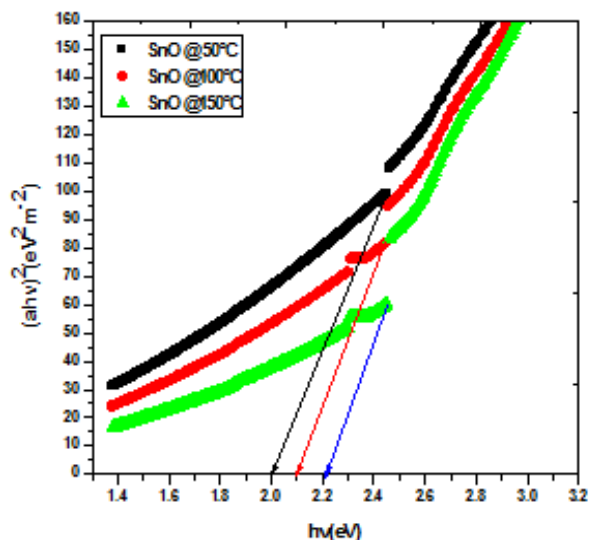


Fig.10.  $(\alpha h\nu)^2$  ( $\text{eV}^2\text{m}^{-2}$ ) Vs  $h\nu$ (eV) for SnO (undoped)

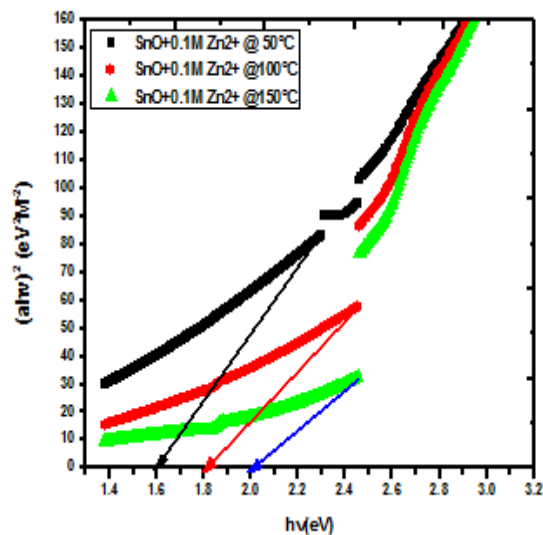


Fig.11.  $(\alpha h\nu)^2$  ( $\text{eV}^2\text{m}^{-2}$ ) Vs  $h\nu$ (eV) for SnO+0.1M of  $\text{Zn}^{2+}$ (doped)

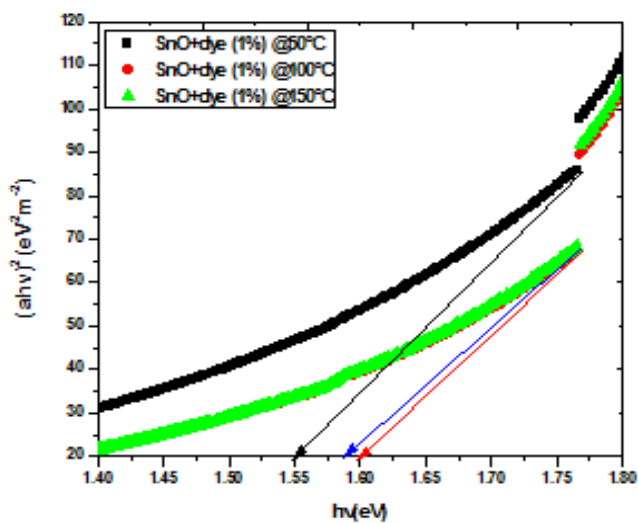


Fig.12.  $(\alpha h\nu)^2$  ( $\text{eV}^2\text{m}^{-2}$ ) Vs  $h\nu$ (eV) for SnO+dye (1%) (doped)

Figures 13 to 15 shows Extin. Coeff. Vs  $h\nu$  (eV) for SnO, SnO:Zn, and SnO:Dye-extract

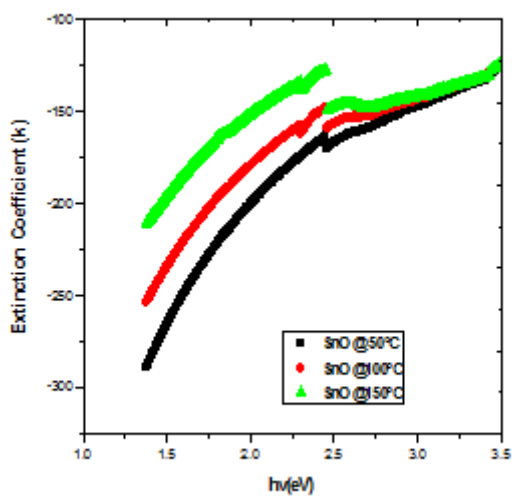


Fig. 13 Extinction Coeff. Vs  $h\nu$  (eV) for SnO (undoped) @50°C, 100°, & 150°C

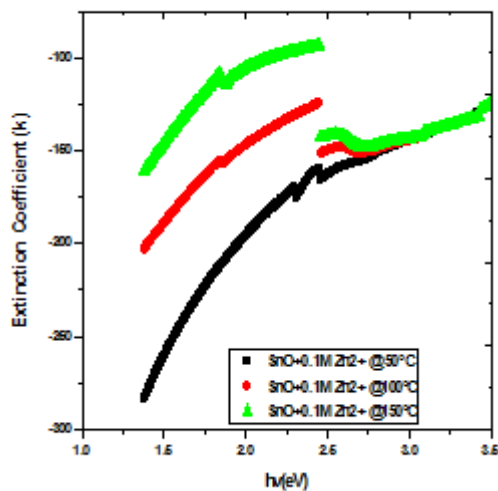


Fig14. Extinction Coeff. Vs  $h\nu$  (eV) for SnO + 0.1M Zn<sup>2+</sup> (doped) @ 50°C, 100°, & 150°C

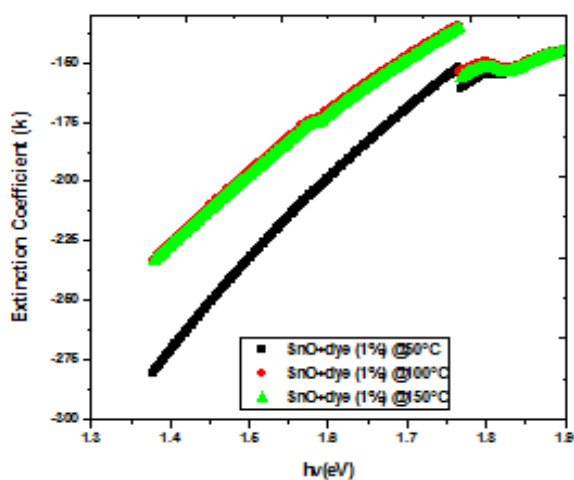


Fig.15 Extinction Coeff. Vs  $h\nu$  (eV) for SnO+dye (1%) (doped) @50°C, 100°, & 150°C

Figures 16 to 18 shows Optical Cond. Vs  $h\nu$ (eV) for SnO, SnO:Zn, and SnO:Dye-extract

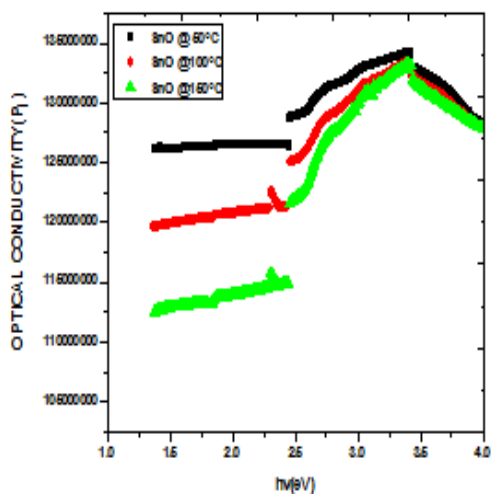


Fig.16: Optical Cond.Vs  $h\nu$ (eV) for SnO (undoped) @ 50°C, 100°, & 150°C

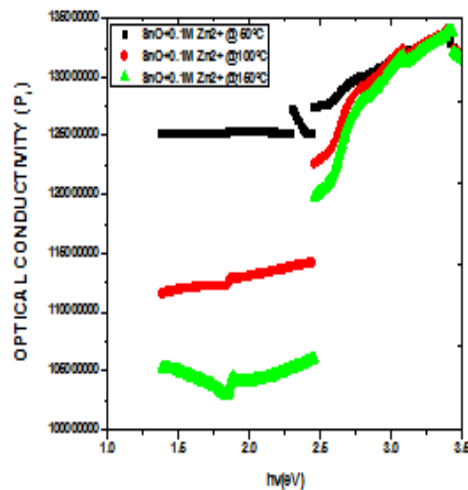


Fig.17: Optical Cond. Vs  $h\nu$ (eV) for SnO + 0.1M Zn<sup>2+</sup>(doped) @ 50°C, 100°, & 150°C

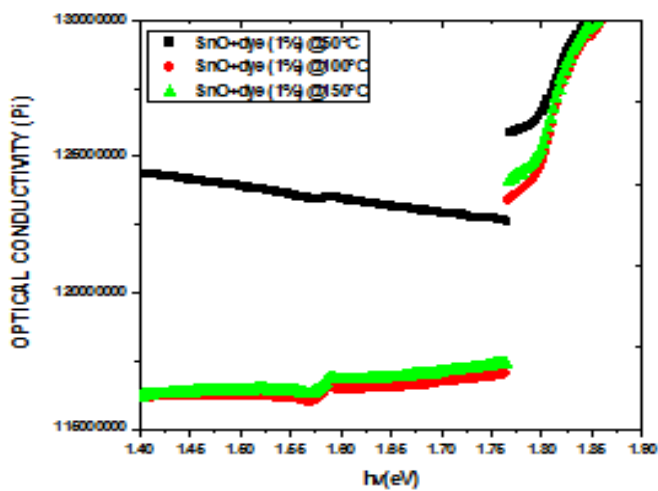


Fig.18: Optical Cond. Vs  $h\nu$ (eV) for SnO +dye (1%) (doped) @ 50°C, 100°, & 150°C



Figures 19 to 21 shows Dielect.Const.( $\epsilon_r$ ) Vs  $h\nu$ (eV) for SnO, SnO:Zn, and SnO:Dye-extract

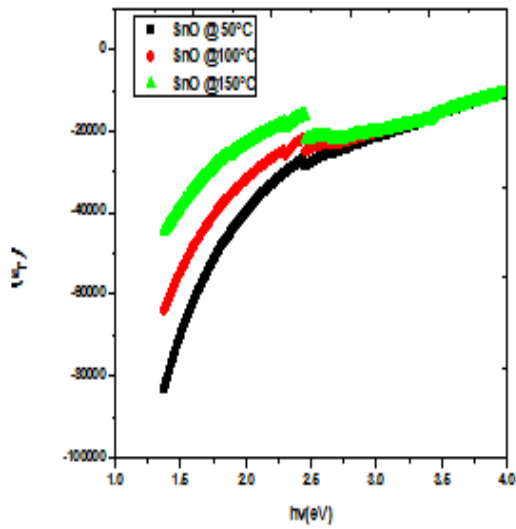


Fig.19: Dielect.Const.( $\epsilon_r$ )Vs  $h\nu$ (eV) for SnO (undoped) @ 50°C, 100°, & 150°C

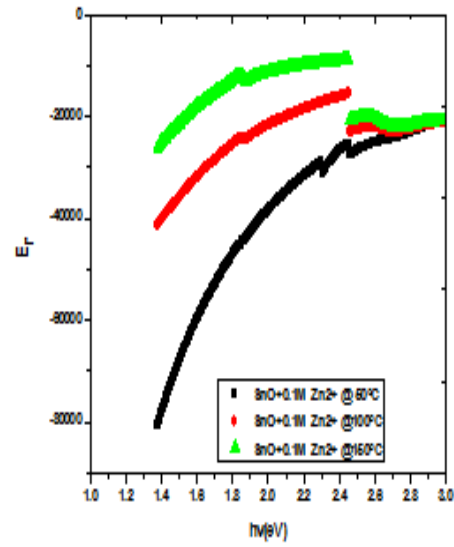


Fig.20: Dielectric Const.( $\epsilon_r$ )Vs  $h\nu$ (eV) for SnO +0.1M Zn<sup>2+</sup>(doped) @ 50°C, 100°, & 150°C

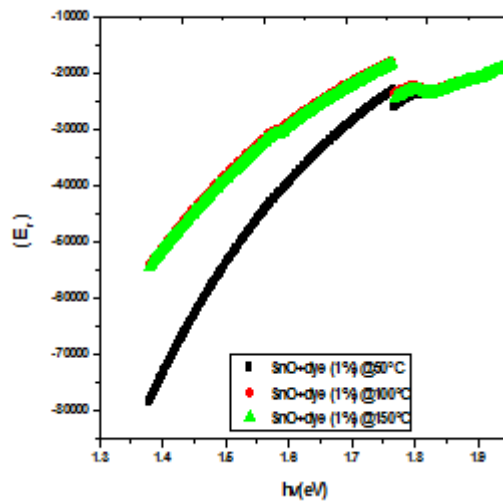


Fig. 21: Dielect Const. ( $\epsilon_r$ ) Vs  $h\nu$ (eV) for SnO + dye(1%) (doped) @ 50°C, 100° & 150°C

Figures 22 to 24 shows Dielect. Const.( $\epsilon_i$ ) Vs  $h\nu$ (eV) for SnO, SnO: Zn, and SnO: Dye-extract

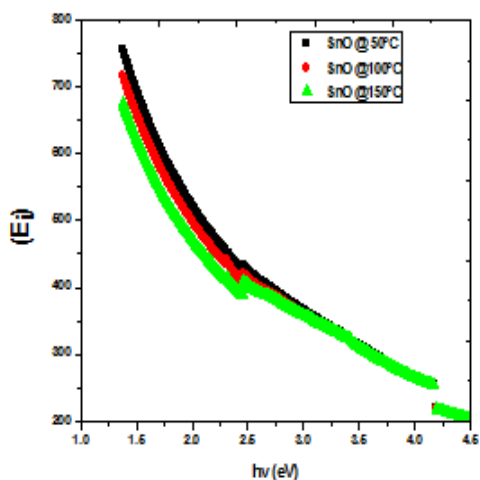


Fig. 22: Dielect. Const.( $\epsilon_i$ ) Vs  $h\nu$ (eV) for SnO (undoped) @ 50°C, 100°, & 150°C

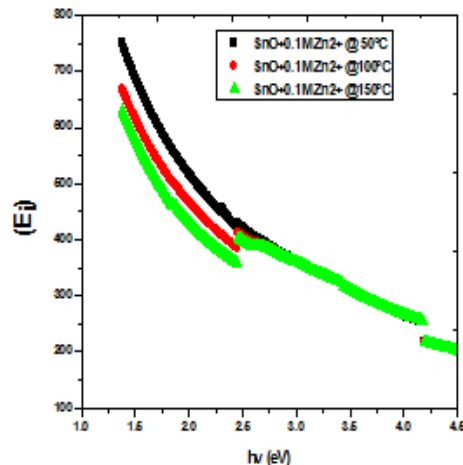


Fig. 23 Dielect. Const.( $\epsilon_i$ ) Vs  $h\nu$ (eV) for SnO +0.1M Zn<sup>2+</sup>(doped) @ 50°C, 100°, & 150°C

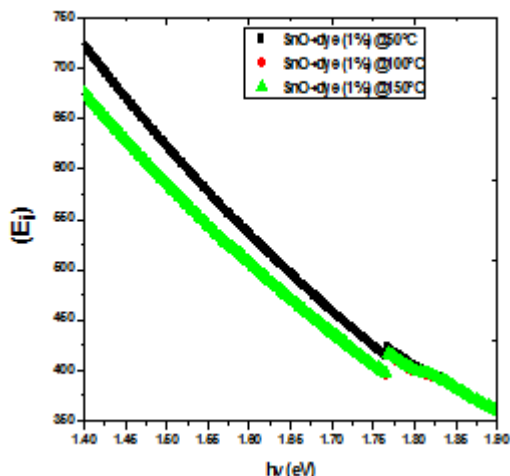


Fig.24: Dielect. Const.( $\epsilon_i$ ) Vs  $h\nu$ (eV) for SnO +dye (1%) (doped) @ 50°C, 100°, & 150°C

**UV-vis-NIR data analysis**

The average transmittance of both un-doped and Zn<sup>2+</sup> doped SnO thin films at 350 nm is above 90 % regardless of the film thickness (Fig. 1-2), which is in good agreement with reported values [14],[15], [5]. The dye doped sample showed an improvement in optical transmission at 625 nm (Fig. 3). In our view, increase in the transmittance could be as a result of the organic content of the dyes. Generally, the required transmittance of transparent conductive thin film solar cells is over 85 % and as such these results indicate that green doped SnO thin films are a good candidate to be used as a window layer in solar cells [5]. The average transmittance of some of the layers both Zn<sup>2+</sup> and dye doped SnO thin films deposited in this work is above 50 % in the visible region. Human eye is sensitive only to the range 400-700 nm and is peaked at 500 nm [2].

This is an important factor in window coatings and is met in some of the samples deposited in this work. The infrared region is the heat portion of

electromagnetic spectrum and some samples of our films showed appreciable transmission in this region, which placed them as suitable materials for the construction of poultry houses for the purpose of admitting heat into the building for warming young chicks that has not developed protective feathers. This has the potential of reducing the cost energy consumption associated with electric bulbs, stoves, etc. These findings are corroborated by the work of other researchers [2], [7], [1]. In the Literature, the transmittance of thin films can be greatly modified by different deposition variables.

The absorbance of the un-doped SnO thin films at various bath temperatures vary from about 0.1-0.7 (Fig. 4). These ranges of values are within the limit stipulated by Lambert-Beer's law (online: <http://www.wikilectures.eu/index-php>). Our values agree with those of Saeideh *et al.* (2011) for SnO thin films but at variance with values obtained by Suresh and Jiban (2015) for SnO<sub>2</sub> thin films all deposited by chemical bath method.

The absorbance generally increases with bath temperature exhibiting a maximum for films deposited at 150°C. The infrared region absorbed better compared to other regions, which is not in agreement with higher absorbance in the UV region reported by Onyia and Nnabuchi (2015), and Florian *et al.* (2019) for chemically deposited SnO thin films subjected to different annealing temperatures. The observed optical behaviour could be attributed to the deposition conditions. The absorbance spectra for 0.1M Zn<sup>2+</sup> doped SnO thin films showed similar trend (Fig. 5) but differed in terms of magnitude of absorption and behaviour of the curves. This clear optical behaviour showed that we can tune the optical properties to suit a specific application by varying growth parameters. When natural dye extract from *tectona grandis* were introduced as dopants, a different behaviour was observed in terms of the absorbance curves and range of values (Fig.6). In our view, the abrupt optical behaviour as a consequence of incorporation of dyes into SnO thin films matrix may be associated with the organic content of the dyes.

The reflectance spectra of all films exhibited a similar trajectory, fluctuating between maxima and minima (Figs.7-9). Peak reflectance can be observed at 350 nm for un-doped and Zn<sup>2+</sup> doped samples of SnO thin films while peak reflectance was observed at 625 nm for dye decorated samples. It can be inferred that the incorporation of Zn<sup>2+</sup> concentrations into SnO matrix did not significantly affect the reflectance. The same trend was observed when the dyes were introduced into SnO films except for 5% dye sample which showed significant increase in optical reflectance. For the Zn<sup>2+</sup> doped SnO films, the lowest reflectance is about 5 % and 25 % for dye decorated layers. This suggests that these films layers can be used as anti-reflective surfaces.

The direct band gap was extracted by linear extrapolation of  $\alpha^2$  versus photon energy plot to zero on the energy axis (Fig. 10-12) shows that the band gap of un-doped SnO films at 50°C, 100°C and 150°C bath temperatures are 2.00 eV, 2.10 eV and 2.20 eV respectively. The band gap for 0.1M Zn<sup>2+</sup> doped layers are 1.60 eV, 1.80 eV and 2.00 eV at bath temperatures of 50°C, 100°C and 150°C respectively. In the configurations, 0.1M of Zn<sup>2+</sup> doped sample showed clear trend with substrate temperature variations. When the dopant was changed to natural dye extracts from *tectona grand* the band gap was greatly adjusted. The band gap for 1 % doped dye samples at 50°C, 100°C and 150°C bath temperatures are 1.55 eV, 1.58 eV and 1.60 eV respectively.

It can be observed that the band gap of the dye doped sample is lower than those of the Zn<sup>2+</sup> doped sample. The narrowing of the band gap may be a consequence

of the organic content of the dyes which were incorporated into SnO<sub>2</sub> films matrix. The incorporation of the dyes shifted the fundamental absorption edge of the un-doped SnO thin films thus providing tuning effect of the band gap for specific applications. Generally, the wide direct band gap energy exhibited by these films make them ideal for use as window layer in hetero-junction solar cells.

The extinction coefficient (k) of the un-doped SnO thin films increased with bath temperature (Fig. 13), which differed in magnitude with those of Saturi *et al.* (2007) for SnO thin films deposited by radio frequency sputtering technique. Islam and Podder (2009) reported extinction coefficient decreases with increasing substrate temperature. The deposition conditions may have accounted for the disparity in extinction coefficient behaviours. When doped with 0.1M Zn-ion concentration, the extinction coefficient showed similar behaviour (Fig. 14). Changing the dopant to dye did not have significantly effect on the extinction coefficient spectra (Figs. 15). The films generally have low extinction coefficient values

The optical conductivity ( $\sigma_{op}$ ) studies for un-doped SnO thin films showed a clear decrease in optical conductivity values with bath temperatures (Fig. 16).

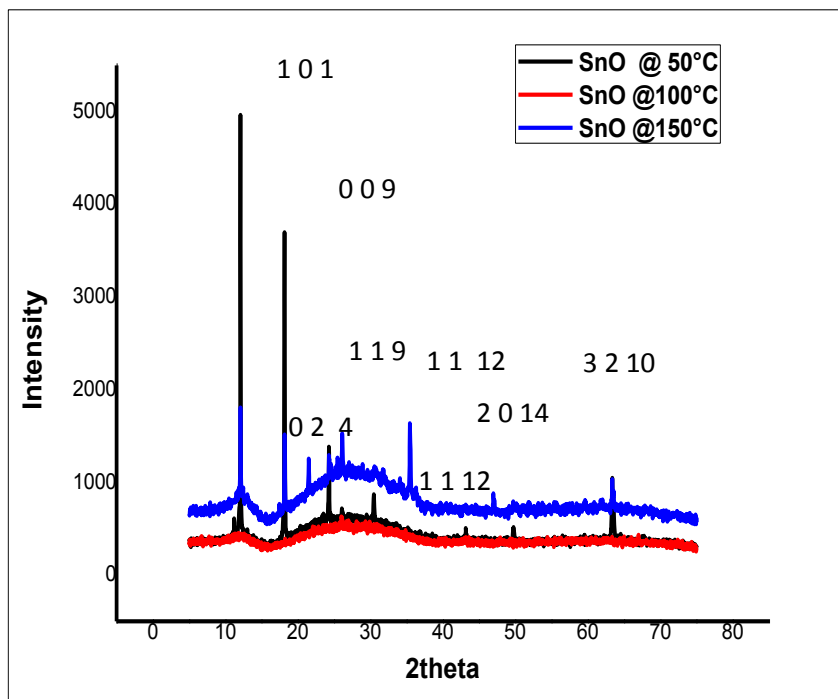
The  $\sigma_{op}$  values reported here are much higher in magnitude compared to reported values in the literature (Augustine *et al.*, 2019). The disparity in  $\sigma_{op}$

values is not unconnected with the deposition conditions. When doped with Zn-ion at 0.1M, no significant difference was observed in terms of the magnitude of  $\sigma_{op}$  values depicted in fig 17.

When dyes were introduced into SnO thin films,  $\sigma_{op}$  spectra showed a different behaviour (Figs.18). Again, the  $\sigma_{op}$  values of the dye coated samples are higher in magnitude when compared with reported values, which may be due to deposition conditions.

The real dielectric constant properties of the SnO thin films were studied for Zn-ion and dye doped samples. The real dielectric constant for un-doped SnO increases with bath temperature between 1.25-2.25eV (Fig.19) while those of 0.1M Zn<sup>2+</sup> did same between 1.30-2.50eV (Fig. 20). When dye was incorporated into the structure, the real dielectric constant spectra depict a similar pattern regardless of the bath temperature.

The imaginary dielectric constant values of the un-doped and Zn-ion doped samples vary in similar pattern. With respect to changes in bath temperatures, un-doped SnO and 0.1M Zn-ion doped samples. This is a clear indication of the influence of growth parameters on the imaginary dielectric constant of the films. the values of  $\epsilon_i$  are higher in magnitude compared to reported values in the Literature (Agbo, 2014; Augustine *et al.*, 2019). Deposition conditions and other growth variables may have accounted for this behaviour.



Figures 25 – 28 represents the X-ray diffraction of the deposited films

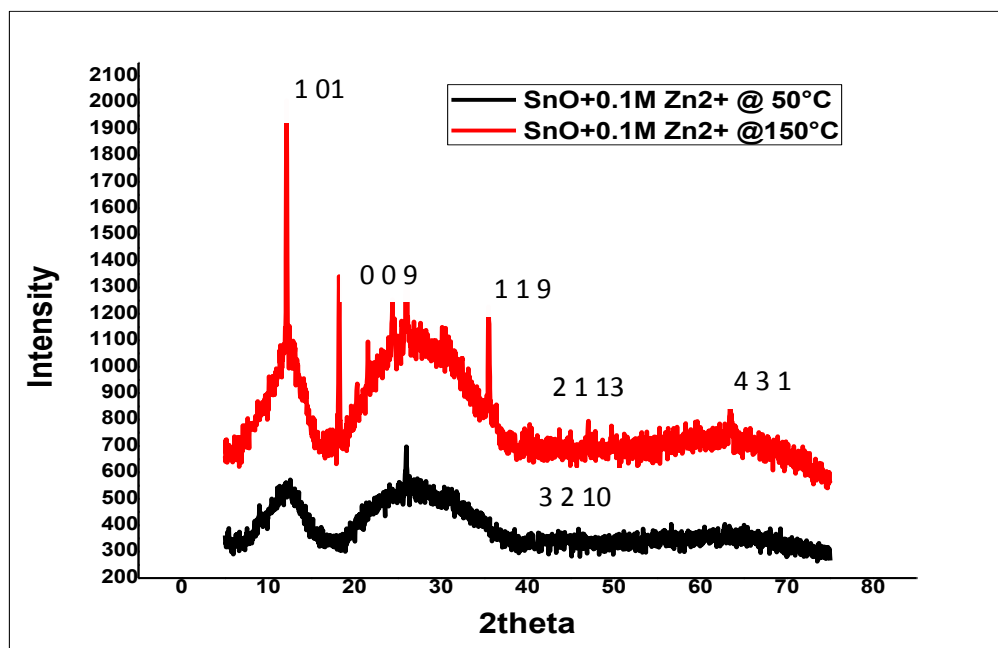


Fig. 26: Intensity Vs 2theta for SnO + 0.1M Zn<sup>2+</sup> @ 50 4 3 1 150°C  
 1 1 4 3 1

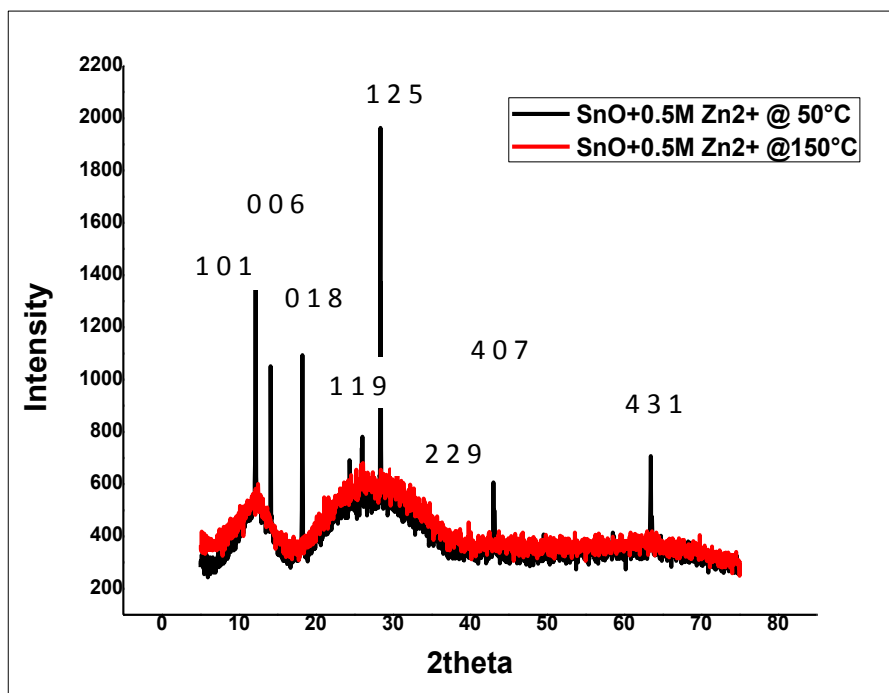


Fig. 27: Intensity Vs 2theta for SnO + 0.5M Zn<sup>2+</sup> @ 50°C, 100°C, & 150°C

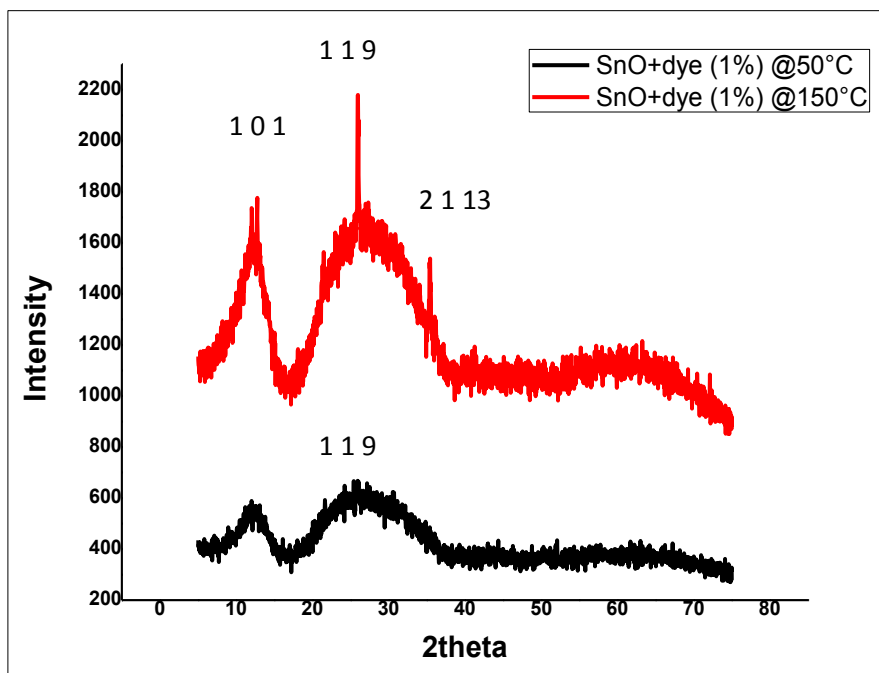


Fig. 28: Intensity Vs 2theta for SnO + (1%) dye @ 50°C, 100°C, & 150°C

#### Structural analysis for SnO: 0.1M Zn<sup>2+</sup> and SnO:0.5M Zn<sup>2+</sup> thin films at different substrate temperatures

The 0.1M Zn<sup>2+</sup> samples were found to exhibit five dominant diffraction peaks with (110), (101), (119), planes while the 0.5M Zn samples were found to exhibit six prominent diffraction peaks with (119), (101), (009), (2 1 13), (3 2 13) and (4 3 1) planes. The

diffractograms of SnO doped Zn are characterized by relatively low intensity for all the samples irrespective of the doping concentration compared to the un-doped SnO samples. The low intensity and a broad diffraction peak were identified, corresponding to an early crystallization of the SnO doped Zn in the oriented structure and such peaks are (101), (009), (119), (2 1 13) and (3 2 10).

This decrease in the peak intensity may be attributed to the increase in the number of defects or reduction in the atomic weight as a result of the substitution of the Sn ion with the Zn ion. The decrease in peak intensity which results to broadness in diffraction peaks might also be due to change in electron density in the conduction band or due to point defect. The crystal structure of SnO did not change because the lattice parameters were not change by the Zn doping, the reason is, Sn<sup>2+</sup> ion have close radii to the Zn<sup>2+</sup> ion, that is; the ionic radius of Sn<sup>2+</sup> is  $r = 0.71 \text{ \AA}$  and Zn<sup>2+</sup> is  $r = 0.74 \text{ \AA}$  (Alpha and Daniel, 2015). This might also be due to the substitution of metal ions in the crystallite structure and therefore result to no structural strain in the SnO:Zn (Alpha and Daniel, 2015). The over-all crystalline quality was found to be decreased due to zinc doping (i.e, the crystallinity of pure SnO is better than that of zinc doped ones). A similar trend of change in preferred orientation has also been observed for Zn:SnO<sub>2</sub> films prepared by ultrasonic spray pyrolysis (Bilgin et al 2004). The experimental peak positions were carefully compared with the standard reported tetragonal SnO, wurtzite ZnO and cubic Zn<sub>2</sub>SnO<sub>4</sub> phases and the miller indices were indexed to the peaks. It has been observed that both SnO and SnO:Zn films possess tetragonal SnO

structure (JCPDS card: 01-088-0287) and no indication for Zn, ZnO and any other mixed phase peaks, which eventually would be expected to form upon mixing SnO with ZnO or vice - versa (Bilgin et al., 2004). The presence of multiple peaks indicate that the films are polycrystalline in nature.

**Structural analysis for SnO:1% dye thin films at different substrate temperatures**

Obviously, the doping of SnO with 1% dye extracts modified the XRD patterns of the un-doped SnO samples. Substrate temperature also affected the structural properties of SnO:dye sample depicting increase in intensity vis-à-vis crystallinity of the films with increase in substrate temperature. The 1% dye sample at substrate temperature 50°C were found to exhibit peaks at 2θ values of 10°, 25.05°, 38.54°, and 73.132° which were assigned to the diffraction lines produced by (119) plane while those of substrate temperature of 150°C exhibit more intense peaks with (101), (119), and (2 1 13) planes. The several multiple peaks as observed in the diffractogram indicate the polycrystalline nature of the films. These findings are corroborated with the report of other authors (Benamet *al.*, 2014; Masheshwari and Karunakarau, 2016; Deya and Hajihashemi, 2019).

The grain size were calculated using scherrer equation;  $D = \frac{k\lambda}{BCOS\theta}$  and the optical density were obtained using  $\sigma = \frac{1}{D}$ .

**Table 1: Average Grain size and Optical Density as a Function of Temperature for SnO**

Temperature (°C)	Average Grain size (nm) (D)	Optical density $\sigma \frac{1}{D^2}$ (nm)
50	2.08398	0.2303
100	1.10123	0.8246
150	1.18568	0.7113

**Table 2: Av. Grain size and Optical Density as a Function of Temp. for SnO:0.1M Zn<sup>2+</sup>**

Temperature (°C)	Average Grain size (nm) (D)	Optical density $\sigma \frac{1}{D^2}$ (nm)
50	1.72892	0.33454
100	0.51878	3.71561

**Table 3: Av. Grain size & Optical density as a Function of Temp. for SnO:0.5M Zn<sup>2+</sup>**

Temperature (°C)	Average Grain size (nm) (D)	Optical density $\sigma \frac{1}{D^2}$ (nm)
50	1.574795	0.403229
100	2.169503	0.212460

**Table 4: Av. Grain size and Optical density as a Function of Temp. for SnO:1% dye**

Temperature (°C)	Average Grain size (nm) (D)	Optical density $\sigma \frac{1}{D^2}$ (nm)
50	2.118833	0.2230
100	2.944093	0.1154

The inter planar d- spacing were deduced using the formula;  $d_{hkl} = a/\sqrt{(h+k+l)^2}$  (nm)

**Table 5: Inter-planar d-spacing (d) as a function of temperature for SnO**

2theta	50°C	100°C	150°
10	7.0848	7.0848	7.0848
18	1.1131	-	1.1131
25	1.0997	1.0997	0.8291
35	-	-	-
40	1.9646	-	-
50	0.9424	-	-
65	-	-	1.964698

**Table 6: Inter-planar d-spacing (d) as a Function of Temperature for SnO:Zn<sup>2+</sup>**

2theta	50°C	100°C	150°
10	7.0848	-	7.0848
18	1.1131	-	1.1131
25	1.0997	-	0.8291
29	2.0036	-	-
38	-	-	-
40	1.9646	-	1.9646

**Table 7: Inter-planar d-spacing (d) as a Function of Temperature for SnO:1% Dye**

2theta	50°C	100°C	150°
10	-	-	7.0848
25	1.0997	-	0.8291

## CONCLUSION

In this study, the effect Zn<sup>2+</sup> and dye extract from green leaves of *tectona grandis* on optical and structural (XRD) properties of the films were examined and analysed. The result showed that the absorbance of the undoped SnO thin films at various bath temperatures vary from about 0.1 – 0.7. The absorbance generally increased with bath temperature exhibiting a maximum for films deposited at 150°C.

The average transmittance of both un-doped and Zn<sup>2+</sup> doped SnO thin films at 350nm is above 90% regardless of the film thickness. The dye doped samples showed an improvement in optical transmission at 625nm. The transmittance increase could be attributed to the organic content of dyes of the green leave of *tectona grandis*. The average transmittance of some of the layers both Zn<sup>2+</sup> and dye doped SnO thin films deposited in this work is above 50% in the visible region.

The reflectance spectra of all films exhibited a similar trend. Peak reflectance was observed at 350nm for un-doped and Zn<sup>2+</sup> doped samples of SnO thin films while peak reflectance can be observed at 625nm for dye decorated samples. It is observed that the incorporation of Zn<sup>2+</sup> concentrations into SnO matrix did not significantly affect the reflectance. It was also observed that the band gap of the dye doped sample is lower: 1.55 – 1.88eV than the Zn<sup>2+</sup> doped sample, 1.60 – 2.22eV. The incorporation of the dyes shifted the fundamental absorption edge of the un-doped SnO thin films thus providing tuning effect of the band gap for solar cells application. The evaluated band

gap showed a red shift upon doping with the energy band gap decreased from 1.60 – 2.22eV to 1.55 – 1.88eV for the investigated doping concentration ranges.

The XRD results reveal that the deposited SnO thin films are polycrystalline in nature having (101) plane as the preferred growth orientation. The diffractograms of SnO doped Zn are characterized by relatively low intensity for all the samples irrespective of the doping concentration compared to the un-doped SnO samples. Obviously, the doping of SnO with 1% dye extracts modified the XRD patterns of the un-doped SnO samples. Substrate temperature also affected the structural properties of SnO:dye sample depicting increase in intensity vis-à-vis crystallinity of the films with increase in substrate temperature. The results equally indicate that there was an increase in the grain sizes that resulted in a decrease in energy badgap of the samples.

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