



Growth and Measurement Of SnO: Zn²⁺ And SnO:Dye Extract from Green Leaves of *Tectonagrandis* Thin Films Deposited Using Spray Pyrolysis Techniques

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ABSTRACT

Dye-sensitized solar cell has a number of attractive features; it provides flexible solar modules, and can be fabricated using conventional method. Dye-sensitized Nanohybrid Zn_xSn_yO_z thin films were prepared on glass substrates using Master Airbrush Brand-model G25, G255-SET and G266-SET to deposit SnO, SnO: Zn²⁺ and SnO. The concentration of the dopant was varied from 0.1M – 0.5M of Zn²⁺ and 1% - 5% of dye-extract. The effect of Zn²⁺ and dye of *Tectonagrandis* on optical and structural 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 0.1–0.7. The dye doped samples showed an improvement in optical transmission at 625nm. The increase in transmittance could be attributed to the organic content of dye. Peak reflectance was observed at 350nm for undoped and Zn²⁺ doped samples. The band gap energy of the dye doped sample is lower ranging from: 1.55 – 1.88eV than that of the Zn²⁺ doped samples, 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 enormous growth of interest in solar energy is the consequence of the need to produce a cleaner energy that is inexpensive and less pollutant to the environment. The solar energy is the most abundant and environmentally friendly renewable energy resource. Photovoltaic's offers an alternative means of producing essential electrical power without further endangering the delicate balance of our fragile ecosystems. It is the direct means of conversion of solar energy to electricity.

Keywords: Green of leave of *Tectonagrandis*, Dopants, Substrate temperature, and Band gap Energy

1.0 INTRODUCTION

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. Given these developments DSSCs hold considerable promise for applications in flexible electronics, smart windows, and indoor photovoltaics, where their unique properties outperform conventional silicon cells (Kokkonen et al, 2021). However, challenges related to long-term stability-especially of liquid electrolytes and natural dyes-remain a focus of ongoing research, with efforts directed at solid state electrolytes and improved encapsulation methods. (Chen et al, 2021).

Recent advancements have focused on improving the efficiency and stability of DSSCs through novel sensitizer designs advanced redox electrolytes, and improved photo anode materials. For example the use of co-sensitization strategies where multiple dyes with complementary absorption spectra are combined has led to power conversion efficiencies exceeding 15% under standard illumination and over 28% under ambient light conditions. This represents a significant improvement over tradition single dye systems. (EPFL Research Group 2022).

Tectonagrandis leaf extracts have also been used as natural dyes due to the presence of anthraquinones and other chromophoric compounds. These dyes have shown potential as natural sensitizers in dye-sensitized solar cell (DSSCs) (Alam et al, 2022). In addition to natural dyes, research has explored modifying semiconductor materials to enhance charge transport and electron mobility. Metal oxides such as tin oxide (SnO) and Zinc oxide (ZnO) are considered potential alternatives to TiO₂ due to their similar conduction band positions and higher electron mobility. (Garg et al, 2021)

In this study SnO, doping is achieved by adding Zn²⁺ atom and dye extract from the leaves of *tectonagrandis* 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 fabricated layers were examined and analysed. The significance of this study includes: It highlights the effects of dye extract of *tectonagrandis* on thin film deposition. It equally x-rays the effect of impurities and temperature on tin oxide thin film properties. This study highlights the pivotal role of impurity and temperature in ensuring better utilization of material in producing cheaper and more efficient thin films of high qualities.

2.0 EXPERIMENTAL DETAILS AND CHARACTERIZATION

The 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 *Tectonagrandis* 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 solution, 40ml of 0.1M aqueous solution of Tin (II) chloride and 4 drops of aqueous ammonia was measured into 50ml glass beaker. The mixtures were stirred thoroughly for 5minutes using magnetic stirrer in order to obtain uniform solution. After stirring, 10ml of the solution were measured out using pump syringe into spray pyrolysis sample bottle that is fixed on a valve rod of air brush and sprayed for 60

seconds on heated substrate at the temperature of 50°C using the compressor at air brush pressure of 22 Pa. This were repeated for 2 different 10ml of the solution deposited at 100°C and 150°C respectively at angle of 28°, height of 36m, distance of 2.5m and time of 60seconds.

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.

3.0 RESULTS

The films fabricated were milky white in nature which gradually turned to brown colour with the increase of temperature. All the deposited films are pore free, uniform and strongly adherent to the slidesurface. For both SnO: Zn^{2+} and SnO-dye composite, Uv-Vis spectroscopy was to determine optical band gap using Tauc plots (Abduh and Al-Odayni, 2023).

3.1 Result of Optical Studies;

The results of optical properties of the deposited film characterized are shown in figures 1 to 24 Figures 1 to 3 show transmittance Vs wavelength for SnO, SnO:Zn, and SnO:Dye-extract

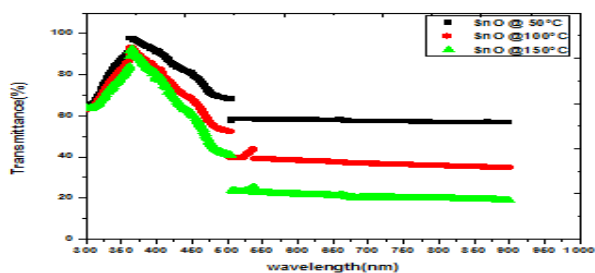


Fig.1. Transmittance Vs Wavelength for SnO (undoped)

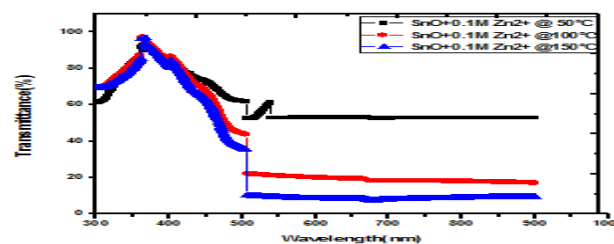


Fig. 2. Transmittance Vs Wavelength for SnO+0.1M of Zn^{2+} (doped)

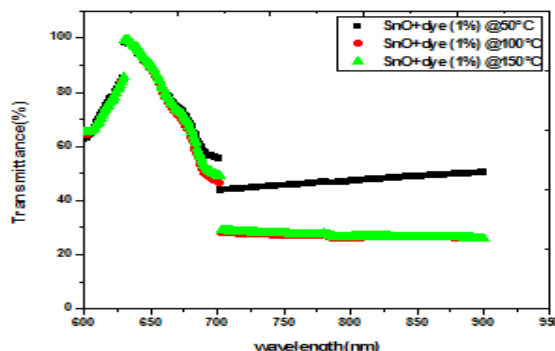


Fig. 3. Transmittance Vs Wavelength for SnO + dye (1%) (doped)

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

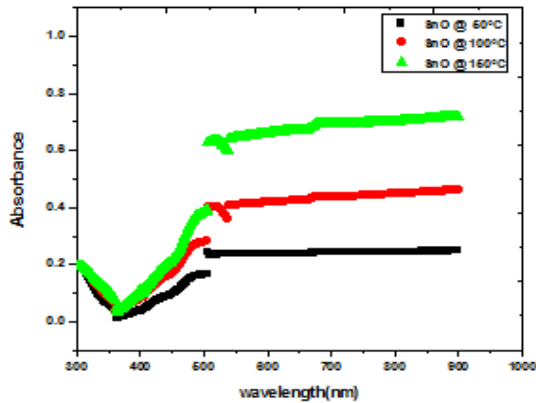


Fig.4. Absorbance Vs Wavelength for SnO (undoped)

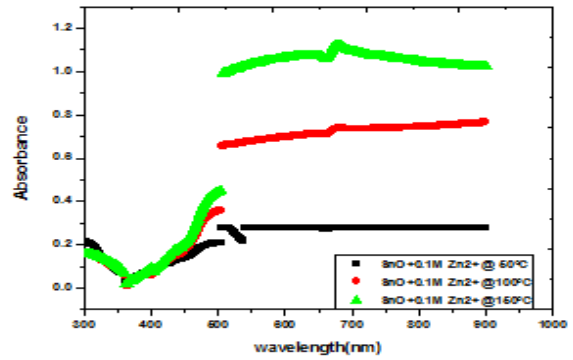


Fig.5. Absorbance Vs Wavelength for SnO+0.1M of Zn²⁺ (doped)

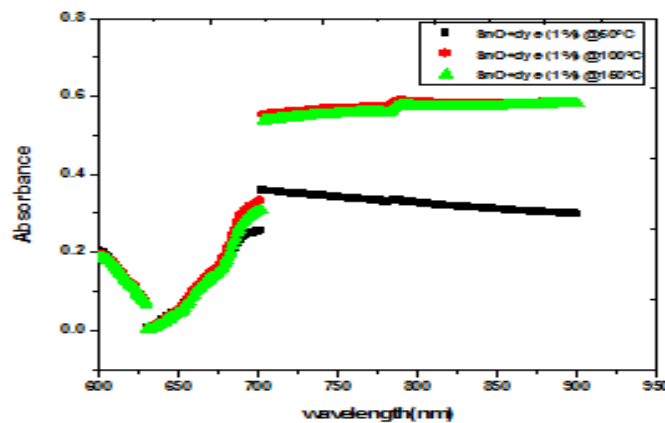


Fig.6. Absorbance Vs Wavelength for SnO+ 1% dye (doped)

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

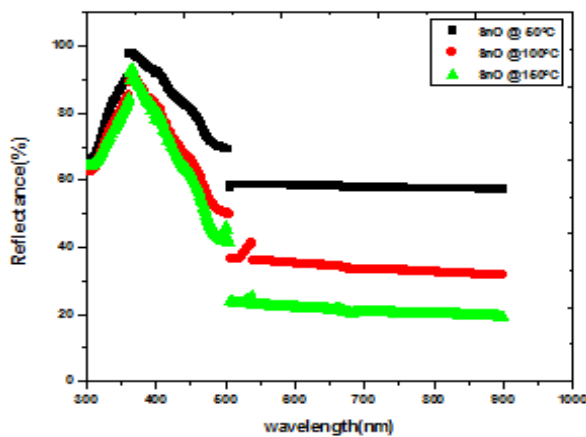


Fig. 7. Reflectance (%) Vs Wavelength for SnO (undoped)

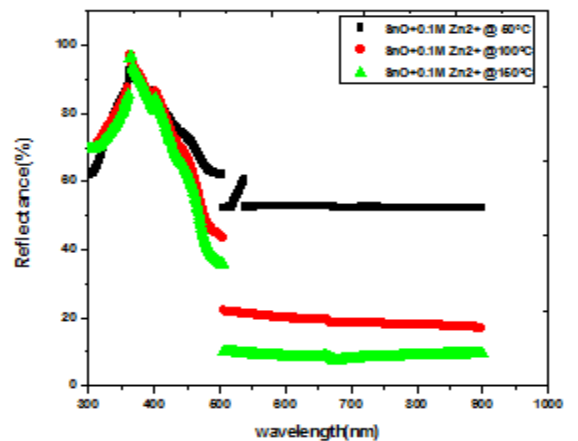


Fig.8. Reflectance (%) Vs Wavelength for SnO+ 0.1of Zn²⁺ (doped)

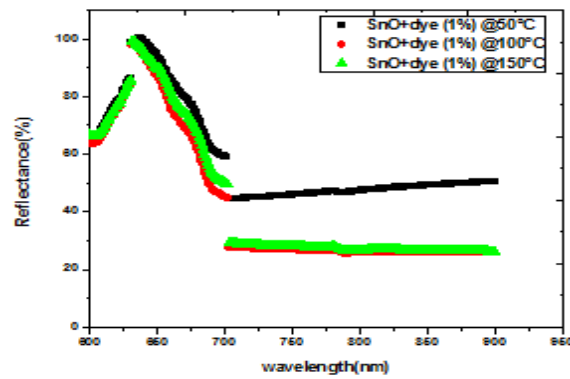


Fig. 9. Reflectance Vs Wavelength for SnO+ dye (1%) dye (doped)

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

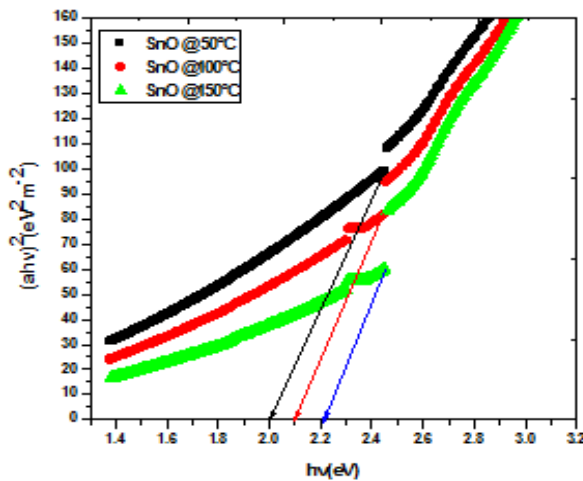


Fig.10. $(\alpha h\nu)^2$ (eV^2m^{-2}) Vs $h\nu$ (eV) for SnO (undoped)

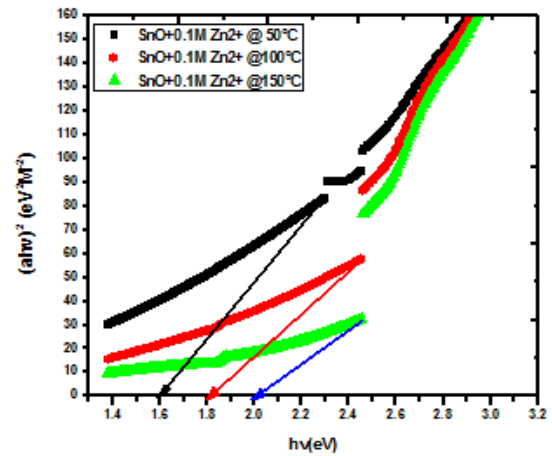


Fig.11. $(\alpha h\nu)^2$ (eV^2m^{-2}) Vs $h\nu$ (eV) for SnO+0.1M of Zn^{2+} (doped)

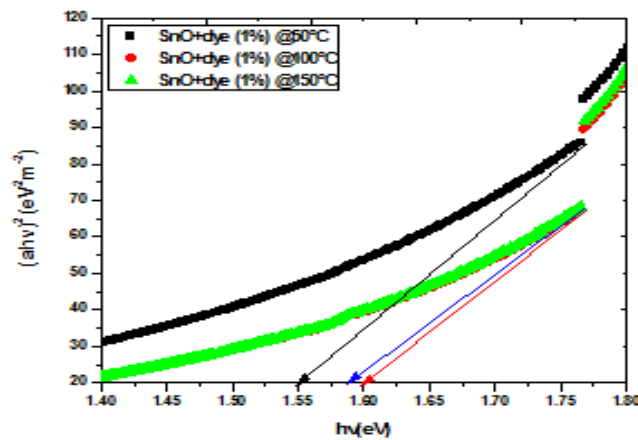


Fig.12. $(\alpha h\nu)^2$ (eV^2m^{-2}) Vs $h\nu$ (eV) for SnO+dye (1%) (doped)

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

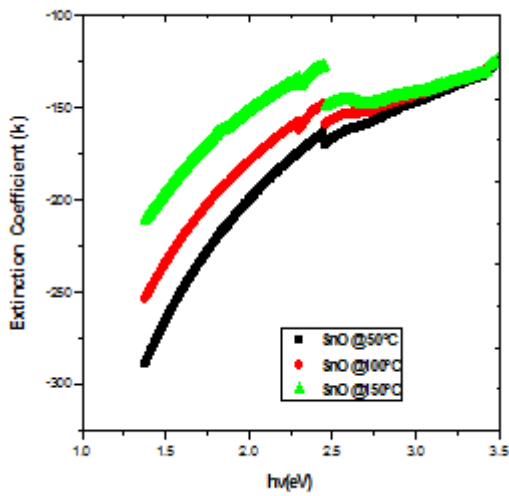


Fig. 13 Extinction Coeff. Vs hv (eV) for SnO (undoped) @ 50°C, 100°, & 150°C

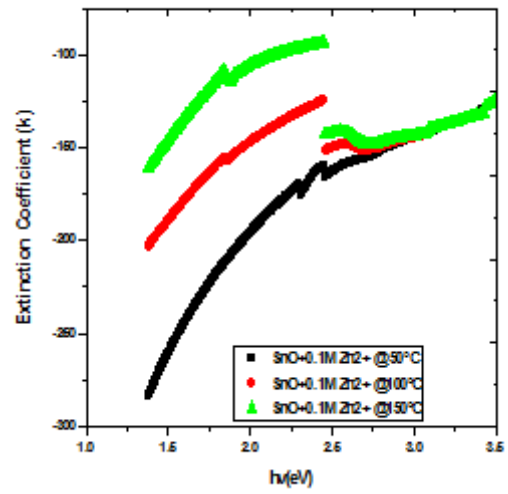


Fig14. Extinction Coeff. Vs hv (eV) for SnO + 0.1M Zn²⁺ (doped) @ 50°C, 100°, & 150°C

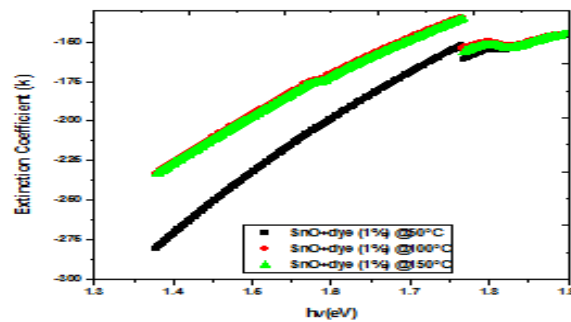


Fig.15 Extinction Coeff. Vs hv (eV) for SnO+dye(1%)(doped) @ 50°C, 100°, & 150°C

Figures 16 to 18 show Optical Cond. Vs hv (eV) for SnO, SnO:Zn, and SnO:Dye-extract

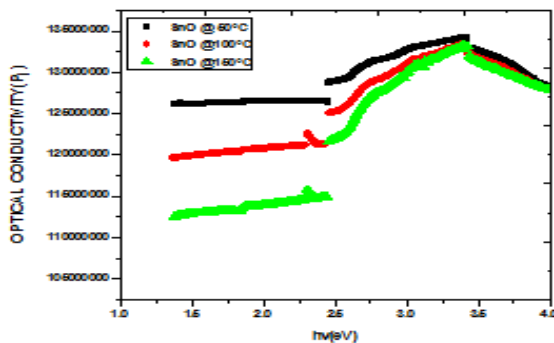


Fig.16: Optical Cond. Vs hv (eV) for SnO (undoped) @ 50°C, 100°, & 150°C

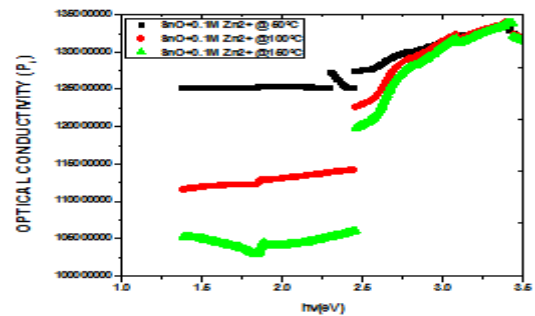


Fig.17: Optical Cond. Vs hv (eV) for SnO + 0.1M Zn²⁺ (doped) @ 50°C, 100°, & 150°C

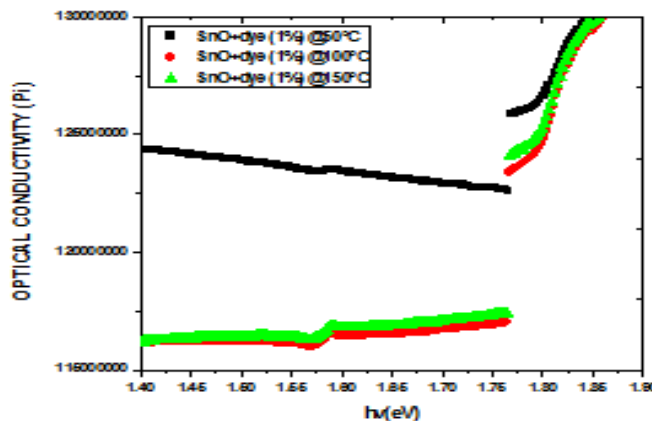


Fig.18: Optical Cond. Vs hv(eV) for SnO+dye (1%) (doped) @ 50°C, 100°,& 150°C

Figures 19 to 21 show Dielect. Const. (ϵ_r) Vs hv(eV) for SnO, SnO:Zn, and SnO:Dye-extract

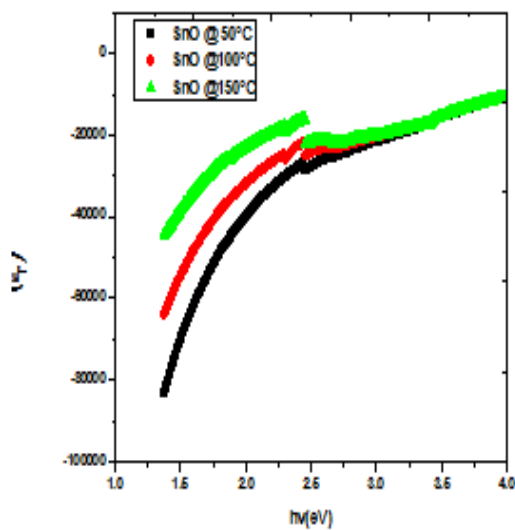


Fig.19: Dielect. Const. (ϵ_r) Vs hv(eV) for SnO (undoped) @ 50°C, 100°, & 150°C

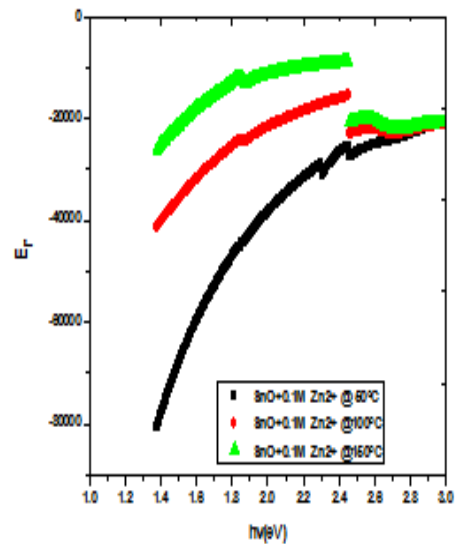


Fig.20: Dielectric Const. (ϵ_r) Vs hv(eV) for SnO +0.1M Zn²⁺(doped) @ 50°C, 100°,& 150°C

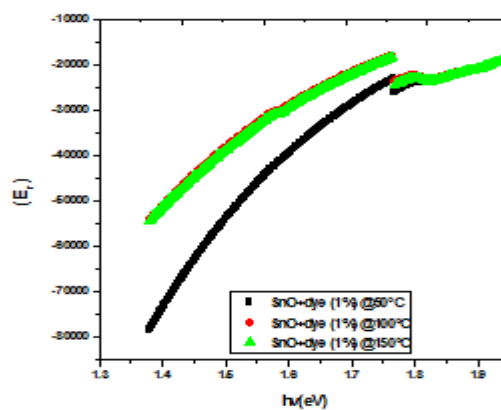


Fig. 21: Dielect Const. (ϵ_r) Vs hv(eV) for SnO + dye(1%) (doped) @ 50°C, 100°& 150°C

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

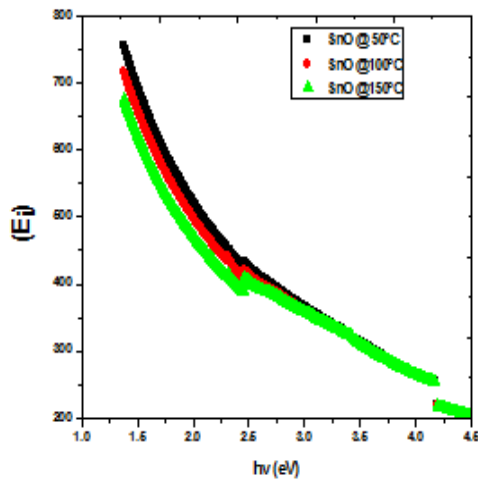


Fig. 22: Dielect Const.(ϵ_i) Vs $h\nu$ (eV) for SnO (undoped) @ 50°C, 100°, & 150°C

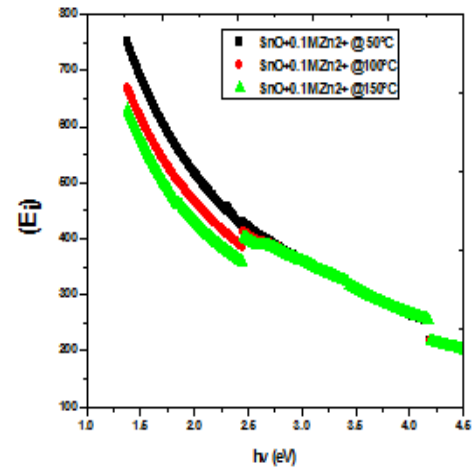


Fig. 23 Dielect. Const.(ϵ_i) Vs $h\nu$ (eV) for SnO +0.1M Zn^{2+} (doped) @ 50°C,

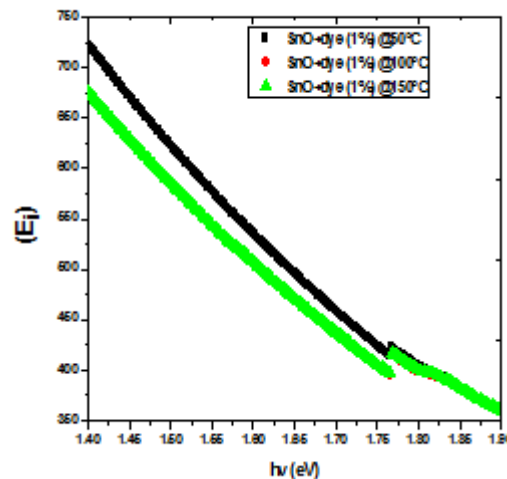


Fig.24: Dielect. Const.(ϵ_i)Vs $h\nu$ (eV) for SnO +dye(1%) (doped) @ 50°C, 100°, & 150°C

4.0 DISCUSSION OF RESULTS

Dye-sensitized thin films fabrication has continued to gain more attention among researchers in thin film technology across the globe due to the fascinating synergetic properties or complimentary behaviours offered by the composite nanostructures. The enormous growth of interest in solar energy is the consequence of the need to produce a cleaner energy that is inexpensive and less pollutant to the environment. The solar energy is the most abundant and environmentally friendly renewable energy resource. Photovoltaic cell offers an alternative means of producing essential electrical power without further endangering the delicate balance of our fragile ecosystems. It is the direct means of conversion of solar energy to electricity. Presently, Silicon base solar cells/panel dominates photovoltaic industries. It is known fact that silicon is an abundant element on earth crust but its high cost of production has hindered wide used of silicon material for

commercial production of electricity. This is because silicon is indirect band gap material and the cost of production is very high. To minimize the high cost of silicon based solar cell, thin film technology solar cells is gaining recognition in recent researches. The optical properties such as absorbance, transmittance, reflectance, energy band gap, extinction coefficient, refractive index, real and imaginary parts of dielectric constant including optical conductivity of the films for un-doped and doped samples of SnO thin films were examined and analyzed. From our result it was that average transmittance of both un-doped and Zn^{2+} 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 [Regan O', Brian, and Gratzel, Michael, 1991], [Rermann K. and Steube M., 1998.], [Deyaet al.,2019]. 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 [Deya, et al.,2019]. The average transmittance of some of the layers both Zn²⁺ 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 [Augustine, et al 2019].

This is an essential 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 [Augustine, C, et al, 2019.], [Fröhlich D., Kenklies R, and Helbig R., 1978.], [Alpha, M. and Daniel, T.O., 2015]. In the Literature, the transmittance of thin films can be greatly modified by different deposition variables.

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 Saeidehet *al.* (2011) for SnO thin films but at variance with values obtained by Suresh and Jiban (2015) for SnO₂ 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 Zn²⁺ 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 tectonagrandis 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 Zn²⁺ 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 Zn²⁺ 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 Zn²⁺ 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 α^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 Zn²⁺ 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 Zn²⁺ 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 Zn²⁺ doped sample. The narrowing of the band gap may be a consequence of the organic content of the dyes which were incorporated into SnO₂ 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 Saturiet *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 (σ_{op}) studies for un-doped SnO thin films showed a clear decrease in optical conductivity values with bath temperatures (Fig. 16). The σ_{op} values reported here are much higher in magnitude compared to reported values in the literature (Augustine *et al.*, 2019). The disparity in σ_{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 σ_{op} values depicted in fig 17.

When dyes were introduced into SnO thin films, σ_{op} spectra showed a different behaviour (Figs.18). Again, the σ_{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 Zn²⁺ 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 (Fig 21). This is a clear indication of the influence of growth parameters on the imaginary dielectric constant of the films. the values of ϵ_i are higher in magnitude compared to reported values in the Literature (Agbo, 2014; Augustine *et al.*, 2019). The extinction coefficient (K) of the un-doped SnO thin films increased with bath temperature. It was equally observed that at doped with 0.1M Zn²⁺ concentration, the extinction coefficient showed similar behaviour and depict opposite trend when doped with 0.2M Zn²⁺. It was observed that when the dopant was changed to dye that no significant effect on extinction coefficient was observed (Fig. 24)

5.0 CONCLUSION

In this study, the effect Zinc ions and dye extract from green leaves of *tectonagrandis* on optical and structural (XRD) properties of the films were investigated 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 Zn²⁺ 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 leaf of *tectonagrandis*. The average transmittance of some of the layers both Zn²⁺ 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 Zn²⁺ 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 Zn²⁺ 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 Zn²⁺ 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 shows 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. The 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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