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Effect of FTO-ITO-/TiO_2/ZnO on Sensitizers in the Fabrication of Dye Sensitized Solar Cell-(DSSC)

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Abstract
Various composite systems have been used on sensitizers for solar cell fabrication for electricity generation. In this study, Fluorine doped tin oxide (FTO), Indium tin oxide (ITO), Titanium oxide (TiO_2) and Zinc oxides (ZnO) were chosen for its outstanding properties in the aspect of increased surface area of absorbtivity and current-voltage performance. Natural dye from Laali
(Lawsonia inermis) stem bark was chosen as the sensitizer. The extracted dye was deposited on these composite systems. The SEM, XRD and EDS were carried out in the solar energy laboratory situated in South Africa. It was observed that ITO emerges the best over FTO while, TiO_2 enhanced the shapes of the dye structure and ZnO enhanced the surface area of absorption.

***Keywords**: FTO: Fluorine doped tin oxide, ITO: Indium tin oxide, ZnO: Zinc Oxide, TiO_2: Titanium oxide, sensitizers and Absorbitivity*

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I. Introduction

Dyes are part of the key elements in DSSCs because they are directly involved in light harvesting, photoelectron generation and electron transfer. For the convectional solar cell, silicon –germanium serves as sensitizers but since its quite expensive to access, local plants have been sourced as an alternative. In order to further enhance the electron transfer composite systems are considered. For this reason, the bonding mechanism and electronic coupling between TiO_2 and dye have been investigated and many dyes with suitable functional-groups have been synthesized to increase the bonding strength between TiO_2 and dyes [1]. Various investigations had been carried out by Rajaram [2] where it was observed the TiO_2 and ZnO are good materials for the enhancement of dye sensitized solar cell fabrication. That ZnO has a large surface area and long conduction pathways that leads straight to the electrode for efficient and fast charge transport and TiO_2 encapsulated the cells. Also, Dong-Joo [3] investigated the photovoltaic performance of transparent conductive indium tin oxide (ITO), titanium indium oxide (ITiO) and fluorine doped tin oxide (FTO) films for dye sensitized solar cell application where the electrical and optical properties such as x-ray diffraction patterns, optical transmittance and photovoltaic characteristics. The near-IR transmittance of ITiO emerged the highest for wavelength over 1.000 nm. TCO glass is made up of FTO Fluorine doped tin oxide and ITO Indium tin oxide conducting glass. This Indium tin oxide coated glass, is electrically conductive and ideal for used in a wide range of devices, including applications such as opto-electronics, touch screen displays, thin film photo-voltaic, energy saving windows, shielding and other electro-optical and insulating applications. ITO glass has the following properties which are sheet resistivity (15 ohm/sq), transmission from 400-700nm wavelength, thickness of the glass about 1.1mm and also the size of the glass can be any size up to 300X300mm while the FTO has a thickness of about 2.2mm, sheet resistivity of 15 ohm/sq, it is a white powder and high in conductivity [4].

Furthermore the photochemical performance of porous ZnO, TiO_2 and bilayer TiO_2/ZnO (TZO) solar cells were investigated by employing five organic dyes. UV-Vis absorption spectra of all dye-sensitized oxide films confirm the anchoring of the dyes with all the ZnO, TiO_2 and TZO films. TZO emerged the best anchoring combination [5]. Based on these literatures, FTO, ITO, TiO_2/ZnO was chosen to enhance good electron transport for light harvesting.

It is widely used in paints, toothpaste and sunscreen. Different morphologies can be synthesized, such as nanoparticles, nanorods, nanowires, nanotubes and nanosheets. TiO_2 has three naturally occurring crystal forms: rutile, anatase and brookite. Rutile is the most thermodynamically stable form, while anatase and brookite can be transformed into rutile form upon heating. Each phase of TiO_2 has different physical properties and thus different functionalities. Rutile TiO_2 scatters white light more efficiently than anatase TiO_2 due to higher refractive index. It is commonly used as white pigments in paints. Anatase is favoured in photo catalysis due to its surface chemistry and larger band gap [6]. The elements of carbon, oxygen, and phosphorus in TiO_2 can function as an electron transfer connector in porous TiO_2 structures. Carbon compounds that form a hexagonal layer of three electrons are used to form a covalent bond with the nearest C atom, while the fourth electron is a free electron that moves through the surface of the layer. This free electron causes the carbon material to conduct electric current [7]. Phosphorus which has five valence electrons is a donor or an n-type doping agent. The fifth electron from the donor is not bound anywhere and can conduct electric current [8], whereas oxygen is an electron acceptor [9], which will capture the electrons produced from dye.

With the presence of carbon, oxygen, and phosphorus elements in TiO_2, the charge from dye can be captured by the molecules of carbon, oxygen, and phosphorus and then can rapidly be passed on to the next TiO_2 particles. This phenomenon causes the charge delivery distance to be shorter so that the electric current can be increased and improve the DSSC performance [10]

Zinc oxide (ZnO) has also been widely investigated due to its wide band gap of 3.37eV at room temperature, which is similar to anatase TiO_2. It can be doped for both n-type and p-type conductivity. Similar to TiO_2, different morphologies can be synthesized using ZnO, such as nanoparticles, nanorods, nanowires, nanotubes, nanoflowers and nanosheets. When compared with TiO_2, the electron mobility of ZnO is much higher, which is an advantage over using TiO_2. ZnO are being applied in UV curing, counterfeit detection, and medical instrumentation, due to its outstanding properties, that is, a wide band gap (3.37 eV), high absorption in the UV range and higher excitonic binding energy (60 meV) than other materials, with similar applications such as GaN (28 meV). Cho [11] also report a simple method for preparing polycrystalline ZnO thin films with good luminescent properties: in which it was observed that the photoluminescence (PL) studies at room temperature for wavelengths between 370 and 675 nm, had a single exciton peak around 390 nm without any deep-level emission and a small PL full width at half maximum (23 meV), indicating that the concentrations of the defects responsible for the deep-level emissions are negligible. Optically pumped lasing action was also observed in these films. The threshold intensity for lasing was ~9 mW/cm^2 [12]. The aim of this study therefore is to investigate the scientific effect of FTO, ITO, TiO_2 and ZnO on the sensitizer laali dye. Sensitizers are dyes from natural plants or synthetic products. Furthermore, stem bark of lawsonia inermis anon edible plant locally available was chosen for this work due to its good anchoring properties.

1.2. Experimental materials

1.21 The Sensitizer-Laali dye

Laali, as popularly called, is grown in the savannah region of West Africa [13] and has the botanical name: *Lawsonia inermis.* It is a very popular natural dye used in colouring fingers, hands, nails and hair (in the northern part of Nigeria). The physiochemical group found in Laali stem bark is called Isoplumbagin, and has chemical structure as shown in figure 1. [13].

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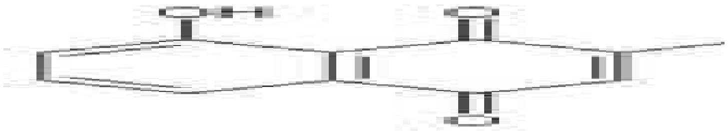


Figure 1: Chemical structure of isoplumbagin dye source: [13]

II. Method

10.30g of Laali stem bark was soaked in 100 ml of ethanol over night. It was further filtered into a beaker; the solution was seen to be greenish in colour (figure 2). When soaked in a powdered form and applied on the skin, it turns brownish red. Here, the TiO_2 was screen printed on the ITO and FTO respectively and later ZnO was sprayed over it producing the ITO/TiO_2/ZnO and FTO/TiO_2/ZnO composite systems. Furthermore the extracted Laali dye solution was deposited on the substrate drop by drop with a syringe. The crystalline ITO/TiO_2/ZnO and FTO/TiO_2/ZnO electrode was examined using x-ray diffraction and optical absorption while the surface morphology of TiO_2 and TiO_2/ZnO films were observed using scanning electron microscopy. Energy dispersive spectrum (EDS) was used to ascertain the atomic compositions.

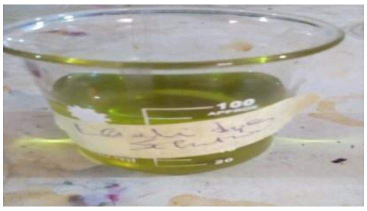


Figure 2: Laali solution in a beaker

(a) (b)

Figure 3: Picture of Slides, (a) FTO and (b) ITO Glasses

Already manufactured FTO and ITO glasses as shown in figure 3, were prepared and sterilized using simple methods. The FTO and ITO glasses were tested to check the polarities using digital multimeter as in figure 4(a) and sterilized by washing in distilled water then in acetone solution and distilled water at intervals (figure 4b) then put in a beaker of distilled water and finally placed inside the ultrasonic cleaner to spin for 15 minutes (figure 4c) . Afterwards solution was poured out and the FTO (a) and ITO (b) rinsed in distilled water and left in the hot air sterilizing cabinet dryer (figure 4d). Pictures of the FTO and ITO is shown in figure 3 (a and b) while the preparation process is shown in figure 4(a-d).

(a) (b) (c) (d)

Figure 4: Testing and sterilizing the FTO and the ITO glass

The preparation of ZnO and TiO_2 substrate is as follows:

A. Zinc acetate powder of 14.4g was weighed and mixed with 100ml of ethanol and stirred rigorously using a magnetic stirrer. Furthermore 0.36 M of sodium hydroxide was prepared and added drop-wise into the solution while still stirring as shown in figure 5 (a and b). Initially it's a clear solution (figure 5(a)) but after adding Sodium hydroxide solution, it turned whitish and sticky as shown in figure 6b and c. This was further centrifuged for 10 minutes to separate the liquid from the particle as shown in picture (d). The particles were placed in a dish and allowed to dry in hot air sterilizing cabinet dryer oven at 60^0C as shown in picture (e). Figure 5(f) shows the dried ZnO particles.

Thereafter 22g of the whitish particle was mixed in 130ml of distilled water and stirred rigorously to allow even mixture. This ZnO solution was used to grow thin film and fabricate the dye sensitized solar cells. A syringe was used to deposit ZnO on the FTO and ITO glass.

Figure 5: Synthesis of ZnO on a magnetic stirrer

B. Furthermore, 6g of powdered TiO_2 was weighed and placed in a mortar for grinding (figure 6a). 9ml of distilled water was added to 0.06ml of acetic acid to get diluted acetic acid. 7.5ml of the prepared diluted acid solution was poured into the powdered TiO_2 inside the mortar, using syringe 20ml each at a time. It was grinded until it forms a paste as shown in figure 6 (b). 1 ml of water with a drop of Tritaniod solution was stirred together rigorously on a separate mortar. Furthermore, this solution was added to the grinded TiO_2 powder to achieve a foamy whitish solution as in (c). The paste of the prepared TiO_2 was now poured into a bottle ready for use as shown in picture (d).The picture of tritaniod solution is as shown in figure 6 (e).

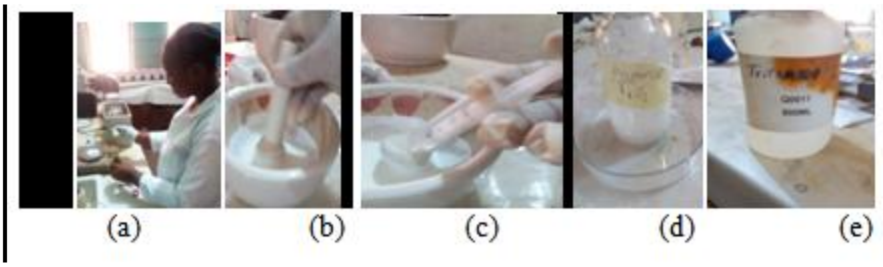


Figure 6: Preparation of TiO_2 paste

2.1 **Deposition Process**

ITO/ZnO/TiO_2/Dye and FTO/ZnO/TiO_2/Dye

The prepared TiO_2 paste was screen printed on the FTO and ITO then annealed at 400^0C for 3 minutes in the furnace for proper adhesion. There after the prepared ZnO solution is sprayed on it using locally made hand spray. Furthermore, the extracted dye solution is injected on it using syringe and annealing is done for proper adhesions. The depositions are as shown in figure 7 (a). the prepared TiO_2/dye, (b) shows TiO_2/ZnO on FTO and (c) the TiO_2/ZnO on ITO glass substrate.

(a) (b) (c)

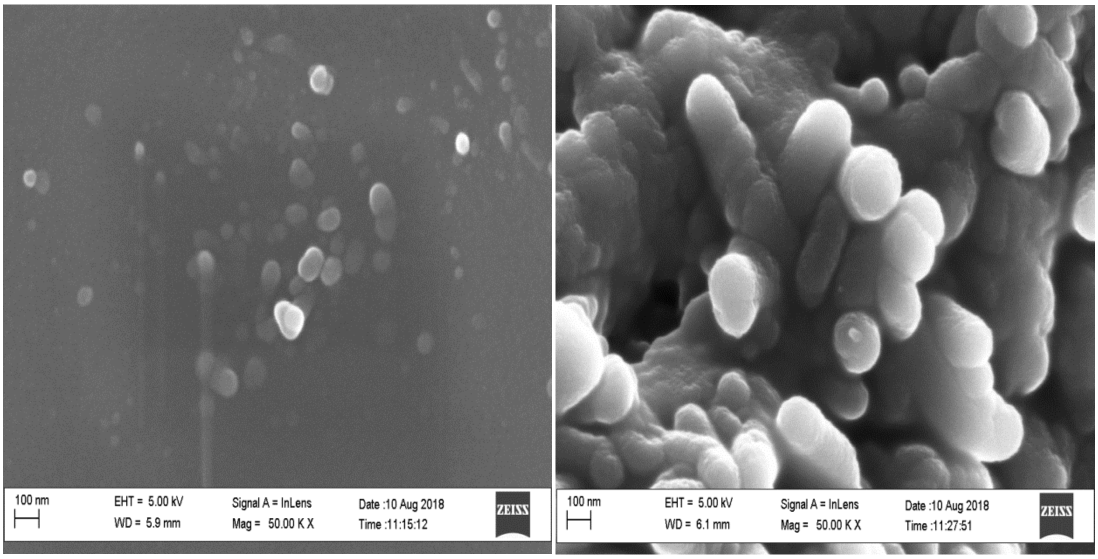
Figure 7: TiO_2/Dye and TiO_2/ZnO FTO and ITO electrode after annealing

III. Experimental Results and Discussion

The SEM, XRD, optical, electronics and EDS results of the Laali dye will be presented to actualize the effect of FTO, ITO, ZnO and TiO_2 on the Laali dye sensitizer.

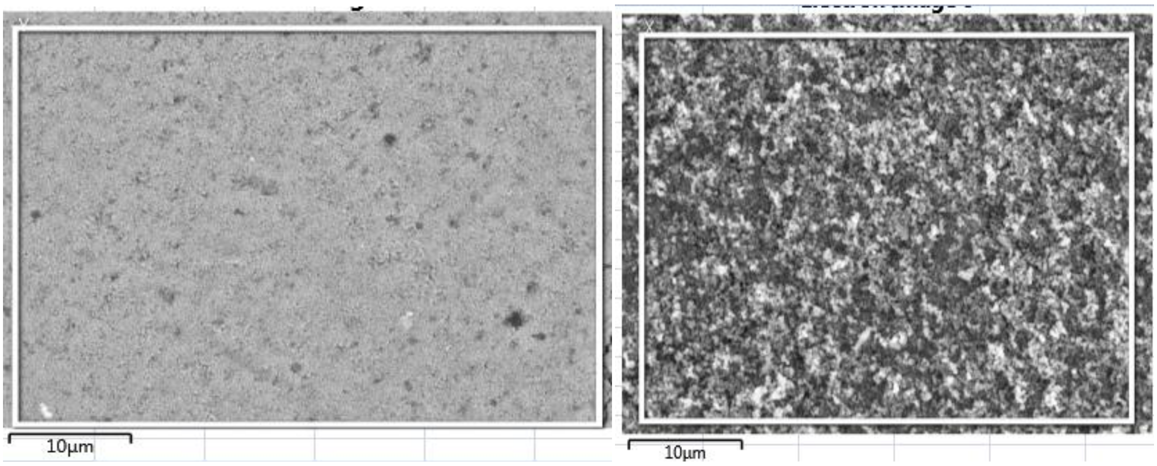
3.1 SEM

Figures 8 to 12, shows the SEM results of Laali dye and its composite systems. Figure 8a and b SEM of laali dye and ZnO/ laali dye in order to ascertain the effect of ZnO on the dye while figure 9 a and b shows FTO/TiO_2/Laali and ITO/TiO_2/Laali dye for wavelength 10 μm to establish the effect of FTO and ITO on the dye. Likewise, Figure 10a and b shows FTO-Laali/ TiO_2 and FTO-Laali/TiO_2/ZnO for wavelength 10 μm to further view the effect of ZnO on both the FTO and the TiO_2 metallic oxide and figure 11 a and b the SEM of ITO-Laali/TiO_2 and ITO/ Laali/TiO_2/ZnO for wavelength 10 μm to examine the effect of ZnO on both ITO and TiO_2 finally, figure 12a and b the SEM of FTO/Laali/TiO_2/ZnO and ITO/Laali/TiO_2/ZnO at wavelength 10 μm to ascertain the effect of FTO and ITO on TiO_2/ZnO metallic oxide for dye sensitized solar cell fabrication. Finally figures 13 and 14 show the SEM of TiO_2 at 50 micro meter and ZnO at 10 micro meters respectively.



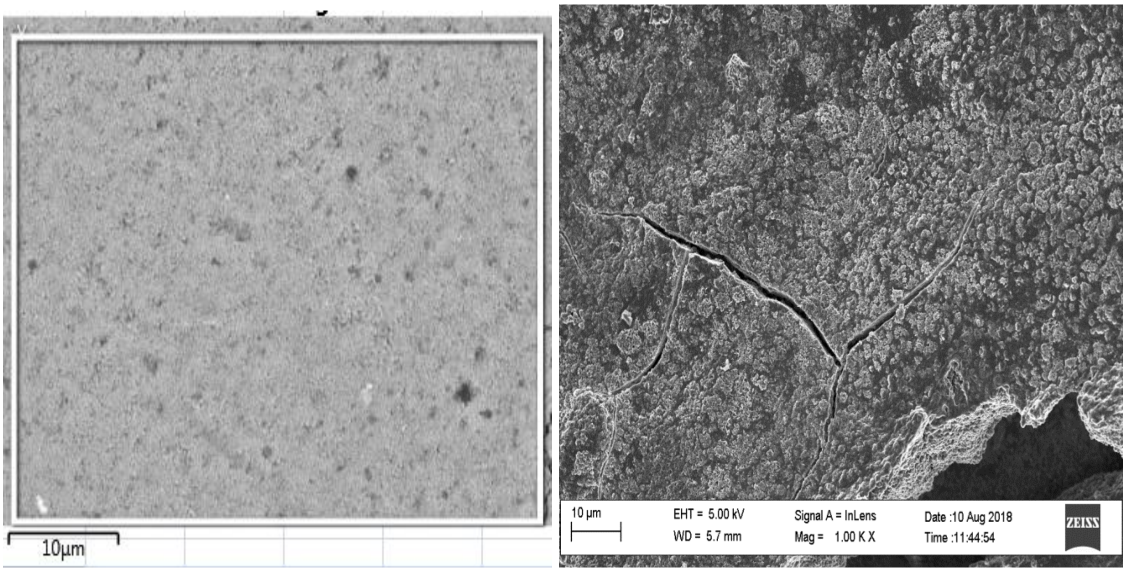
(a) (b)

Figure 8: SEM of Laali dye and ZnO/ Laali at magnification of 50kx for wavelength 100nm



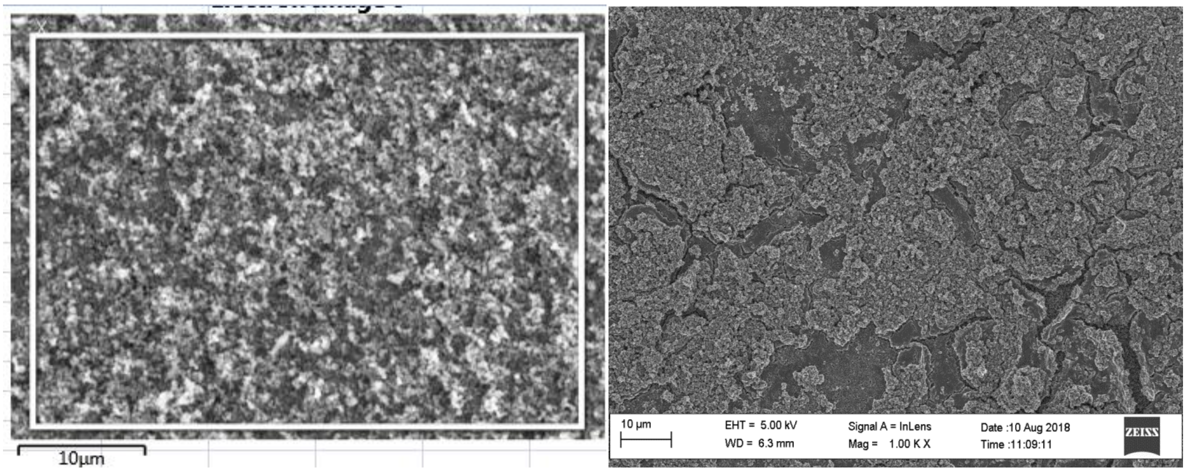
(a) (b)

Figure 9: SEM of FTO / TiO$_2$ /Laali and ITO /TiO$_2$/Laali for wavelength 10 μ m



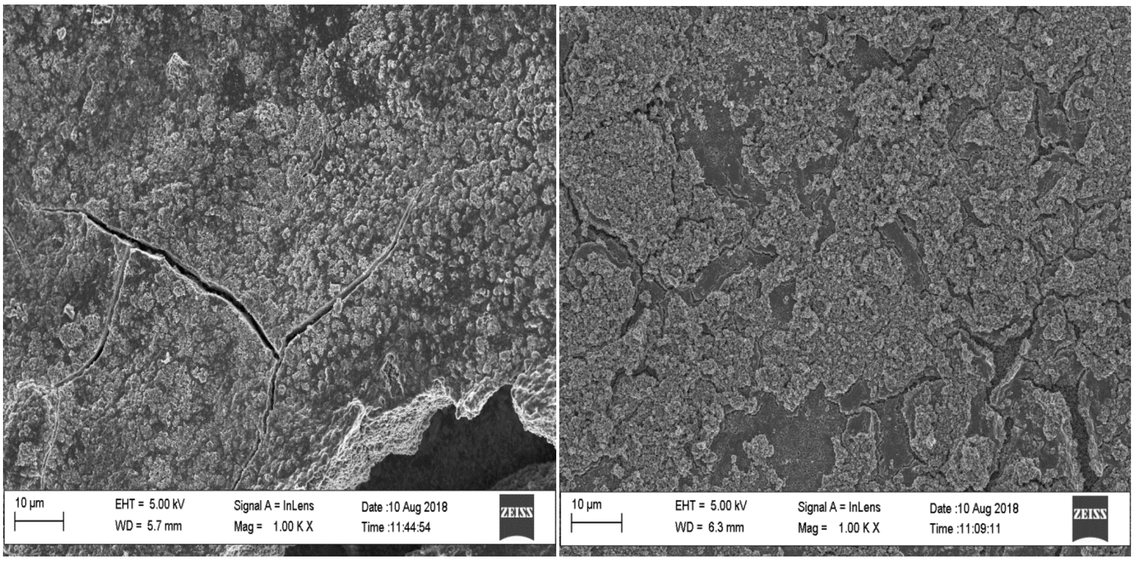
(a) (b)

Figure 10: SEM of FTO / TiO$_2$/Laali and FTO /TiO$_2$/ZnO/Laali for wavelength 10 μ m



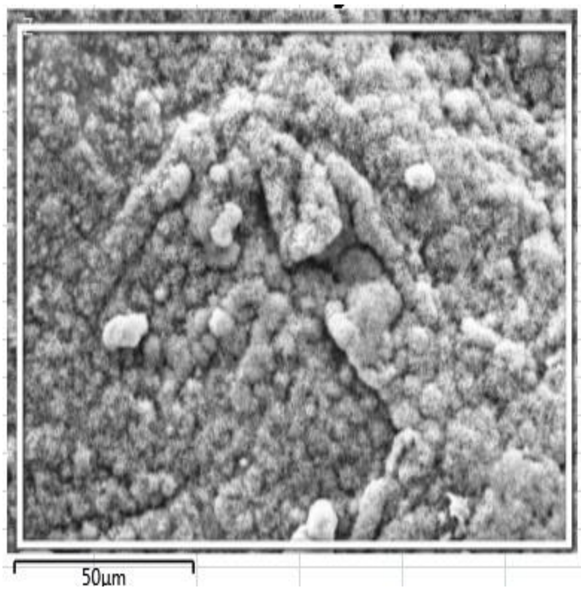
(a) (b)

Figure 11: SEM of ITO /TiO_2/Laali and ITO /TiO_2/ZnO/ Laali for wavelength 10 μ m



(a) (b)

Figure 12: SEM of FTO/TiO_2/ZnO/Laali and ITO /TiO_2/ZnO/Laali at wavelength 10 μ m



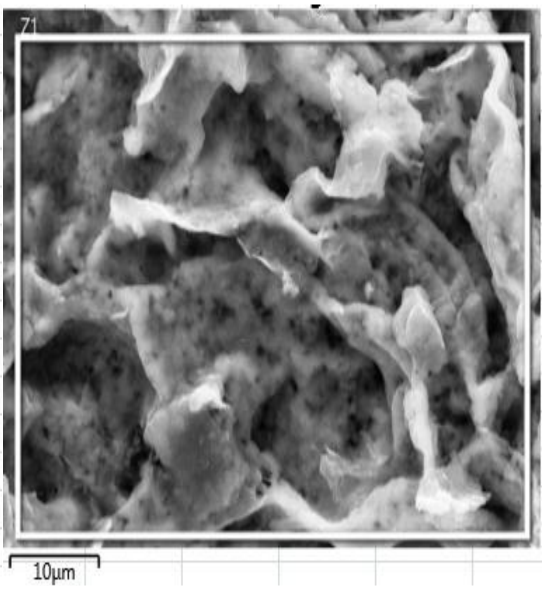


Figure 13: SEM of TiO_2 at 50 μ m wavelength **Figure** 14: SEM of ZnO at 10 μ m wavelength

From the SEM results it was observed that Laali anchored properly on ZnO as shown in figure 8 which agrees with Wasiu [9] research work.

3.2 XRD

Figures 15 to 20 shows the XRD result of Laali composite systems. Figure 15 and 16, the XRD of ZnO/Laali and FTO/ZnO/ Laali to ascertain the effect of FTO while figure 17 the XRD of ITO /TiO_2/Laali and figure 18 the XRD of ITO/ TiO_2/ZnO/ Laali to further view the effect of ZnO. Likewise, figure 19 and 20 of XRD of FTO /TiO_2/ZnO/Laali and ITO /TiO_2/ZnO/ Laali in order to verify the most efficient composite system for dye sensitized solar cell fabrication. Furthermore figures 21 and 22 shows the XRD of TiO_2 and ZnO respectively.

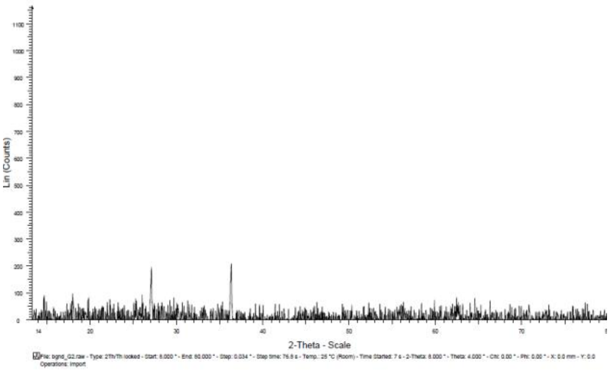


Figure 15: XRD of ZnO/Laali dye

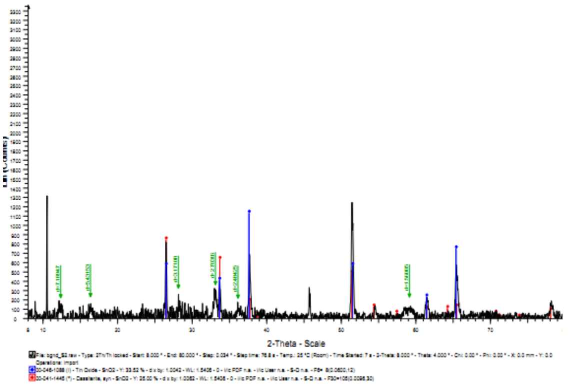


Figure 16: XRD of FTO/ZnO/ Laali dye

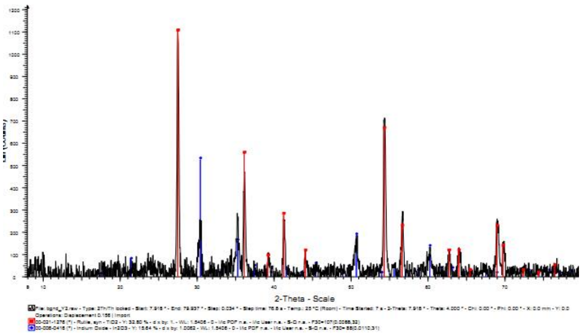


Figure 17: XRD of ITO /TiO_2 /Laali

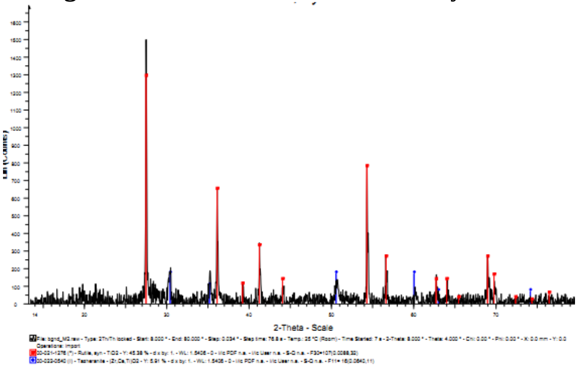


Figure 18: XRD of ITO /TiO_2/ZnO/Laali

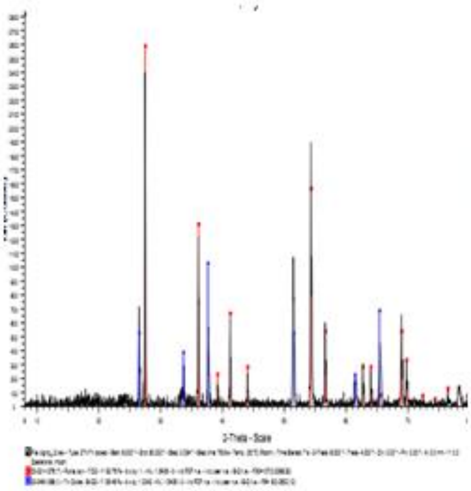


Figure 19: XRD of FTO /TiO_2/ZnO/Laali

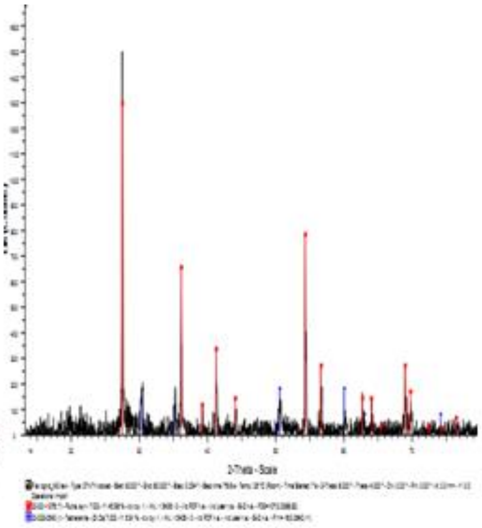


Figure 20: XRD of ITO/TiO_2/ZnO/Laali

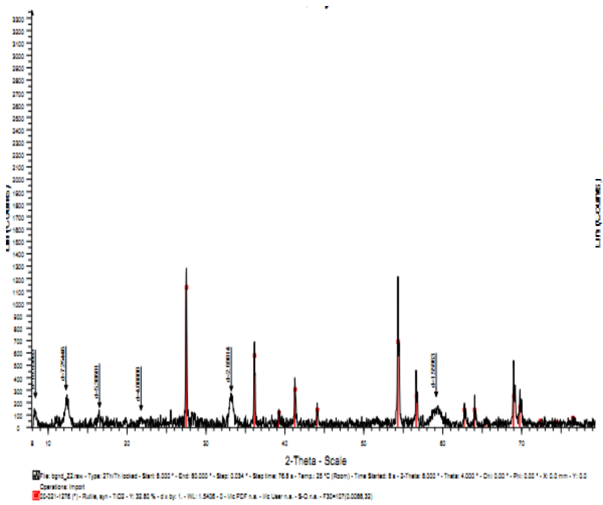


Figure 21: XRD of TiO_2

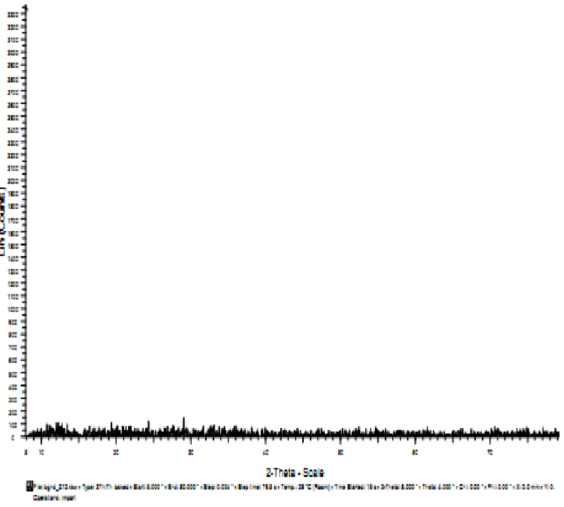
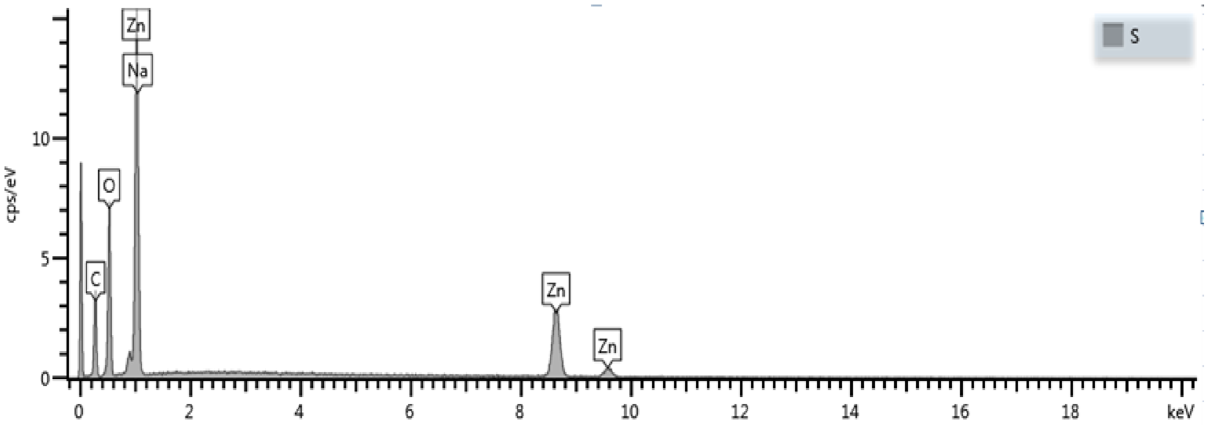


Figure 22: XRD of ZnO

Figures 15-22 shows the XRD pattern of ITO, FTO, TiO_2 and ZnO thin films after annealing at 400^0C unlike Doog-Jo who annealed at 450^0C. [5]. TiO_2 enhances the peaks in the rutile structure .which is in conformity with the general properties of titanium oxides in light harvesting.

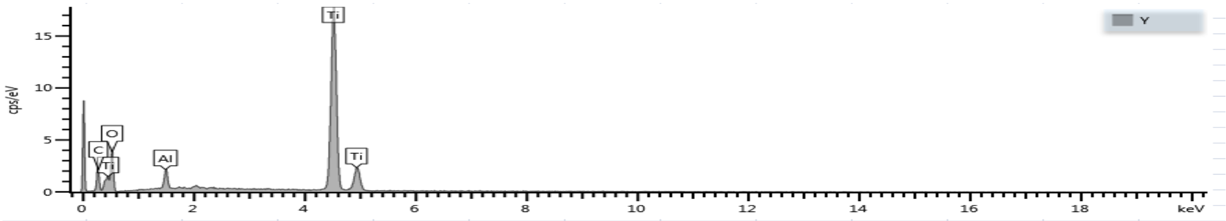
3.3 EDS

Figure 23 to 27 shows the Energy dispersive spectrum of Laali composite system. Figure 23 shows the EDS of FTO/ZnO/Laali and figure 24 the EDS of FTO/TiO_2 /Laali likewise, figure 25 the EDS of ITO/TiO_2/Laali in order to ascertain the different components of the atoms contained in the Laali composite systems. Furthermore, figure 26, shows the EDS of TiO_2 and Figure 27 show the EDS of ZnO to ascertain their atomic composition independently.



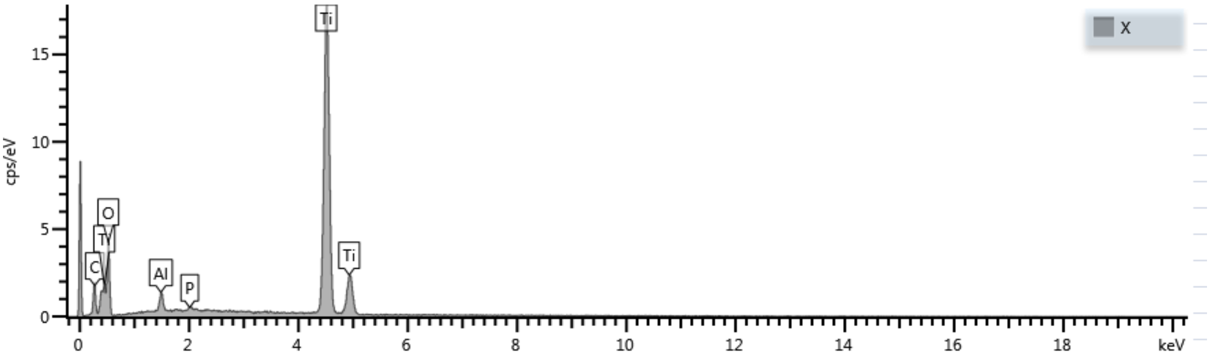
Element	Atomic%
C	51.41
O	30.73
Na	5.24
Zn	12.62
Total:	100

Figure 23: EDS of FTO /ZnO/Laali dye



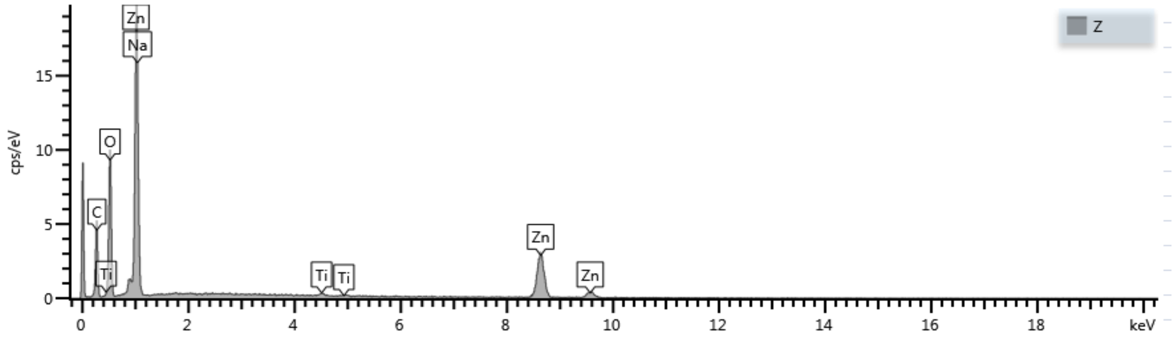
Element	Atomic%
C	24.86
O	47.79
Al	2.07
Ti	25.28
Total:	100

Figure 24: EDS of FTO /TiO$_2$/Laali dye



Element	Atomic%
C	18.77
O	52.75
Al	1.25
P	0.17
Ti	27.06
Total:	100

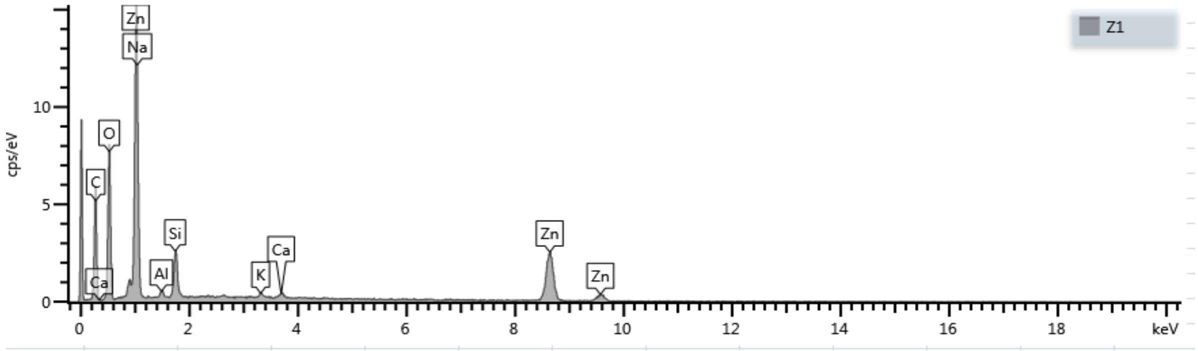
Figure 25: EDS of ITO /TiO$_2$/Laali dye



Element	Atomic%
C	51.84

O	32.46
Na	5.63
Ti	0.15
Zn	9.91
Total:	100

Figure 26: EDS OF Plain glass TiO_2 film



Element	Atomic%
C	57.08
O	28.4
Na	4.04
Al	0.3
Si	1.75
K	0.14
Ca	0.17
Zn	8.12
Total:	100

Figure 27: EDS OF ZnO Film

3.4 Uv-visible (optical characteristics)

The transmittance/Absorbance of TiO_2/ZnO/FTO/ITO is presented in figures 28-36. Figure 28 and figure 29 shows the transmittance of TiO_2 Film and ZnO film respectively while figure 30 and 31 shows the transmittance and absorbance of TiO_2/ZnO film while Figure 32 show the absorbance of ZnO. While figures 33, 34, 35 and 36 shows the transmittance and absorbance of FTO and ITO TCO glasses respectively. Finally figures 37, 38, 39, 40, 41 shows the transmittance of laali dye, the transmittance and absorbance of FTO/TiO_2/ZnO/laali and ITO/TiO_2/ZnO/laali dye

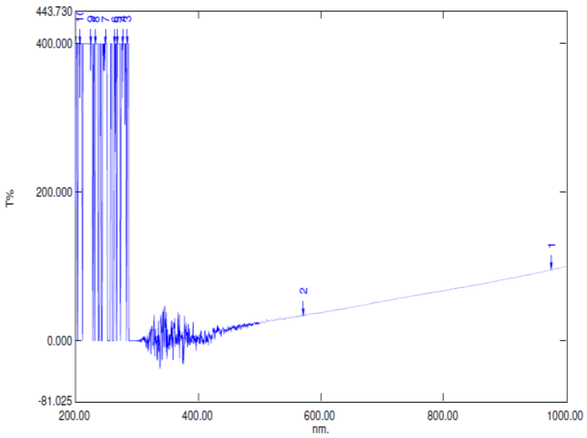


Figure 28: Transmittance of TiO_2 Film

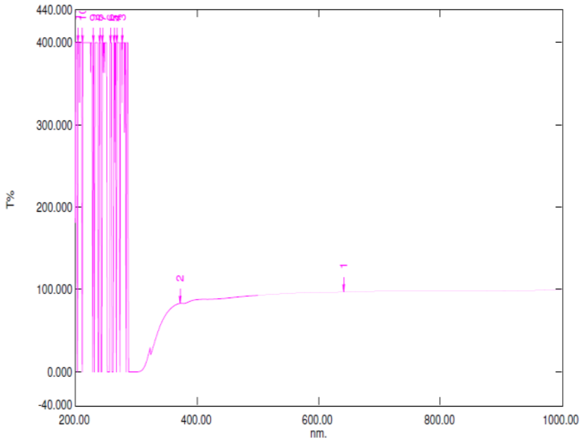


Figure 29: Transmittance of ZnO Film

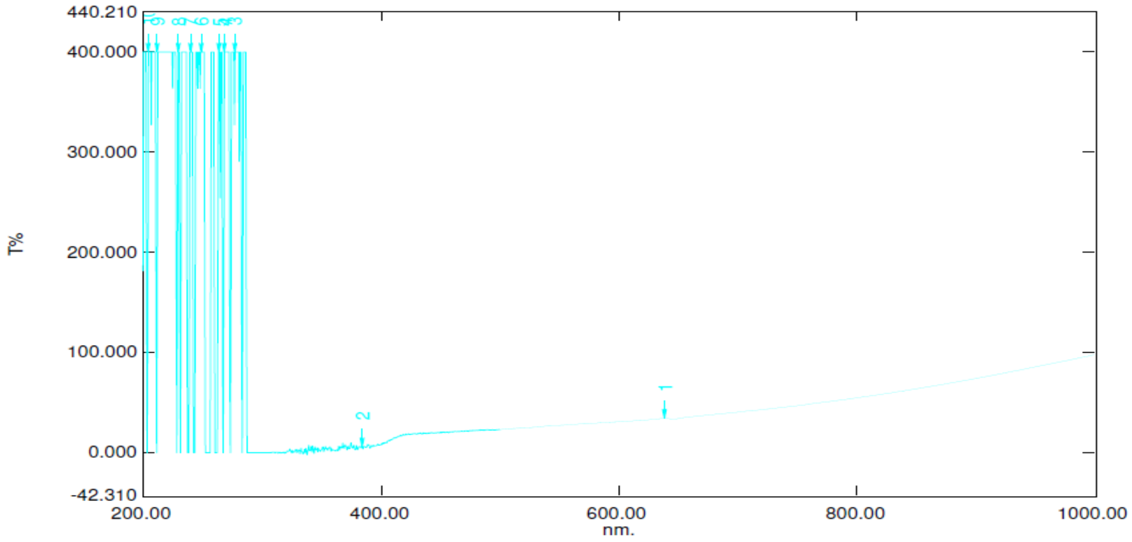


Figure 30: Transmittance of TiO_2/ZnO Film

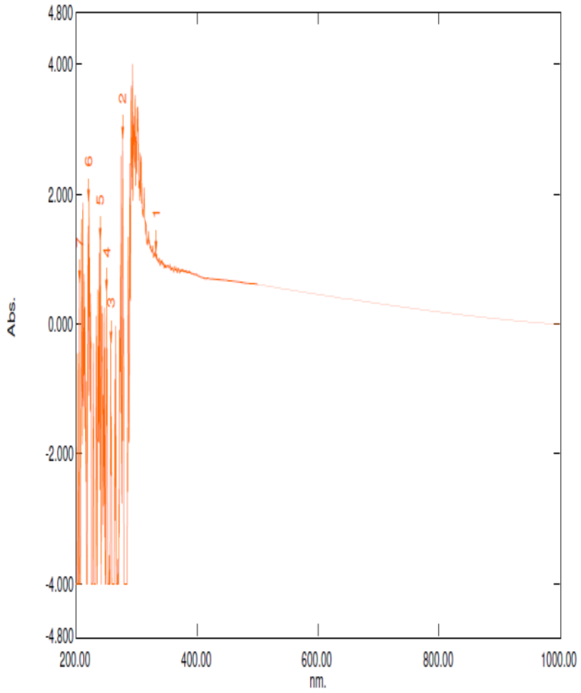


Figure 31: Absorbance of TiO_2/ZnO film

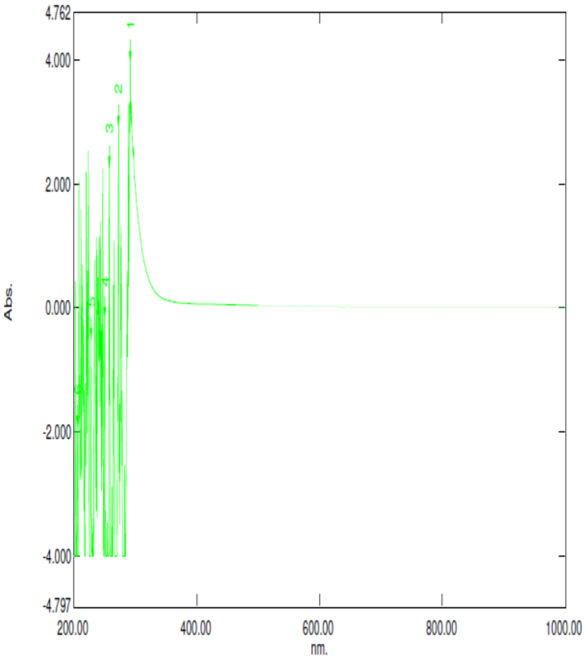


Figure 32: Absorbance of ZnO Film

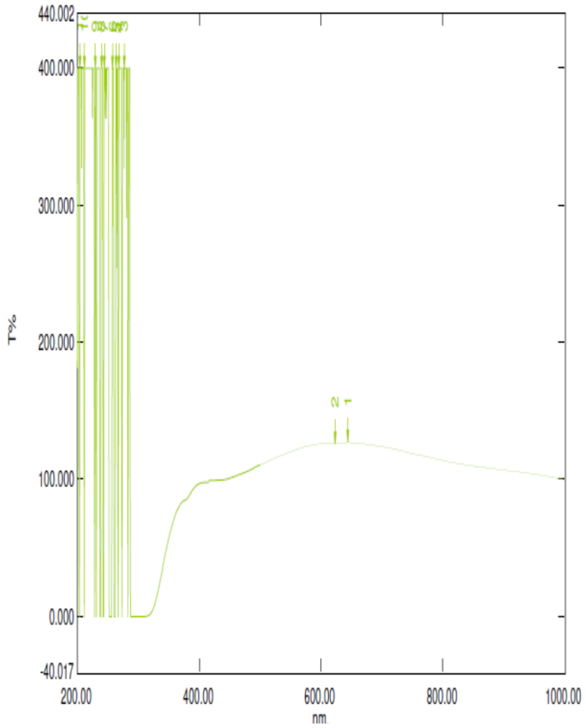


Figure 33: Transmittance of FTO glass

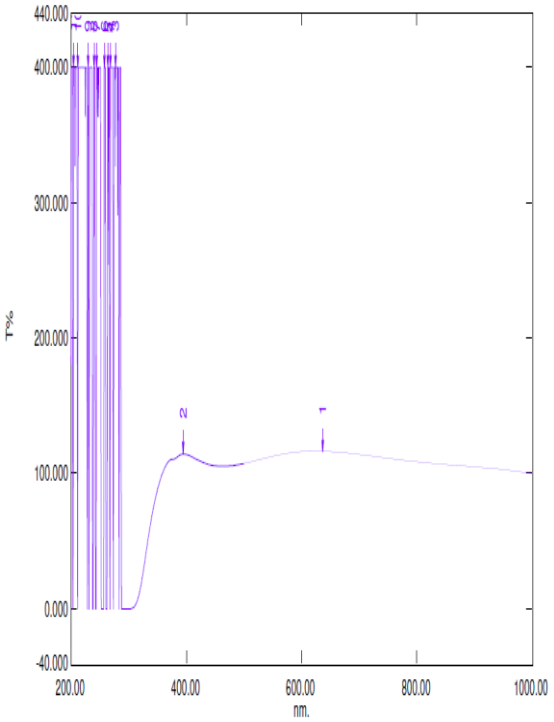


Figure 34: Transmittance of ITO glass

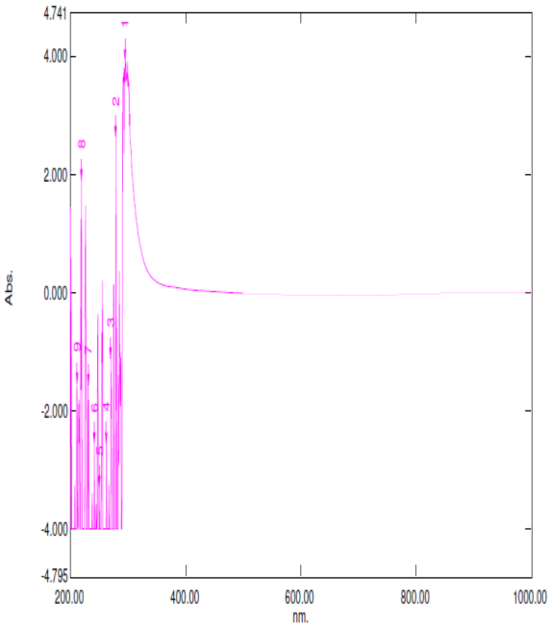


Figure 35: Absorbance of FTO glass

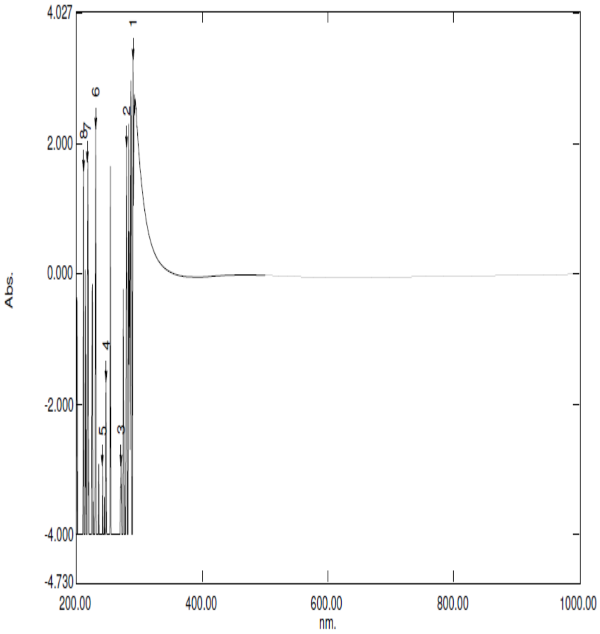


Figure 36: Absorbance of ITO glass

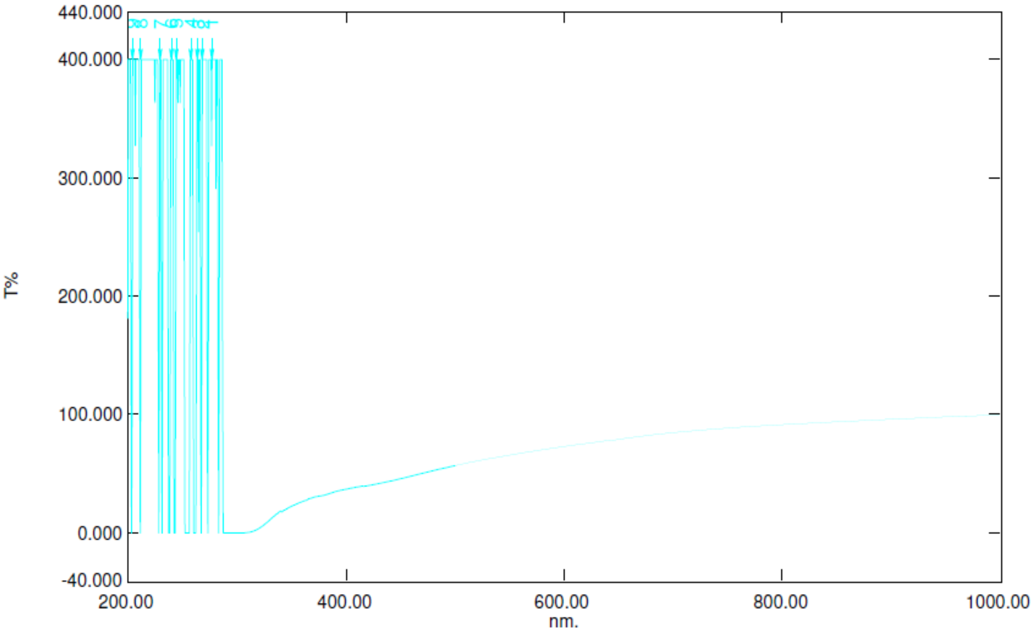


Figure 37: Transmittance of Laali dye

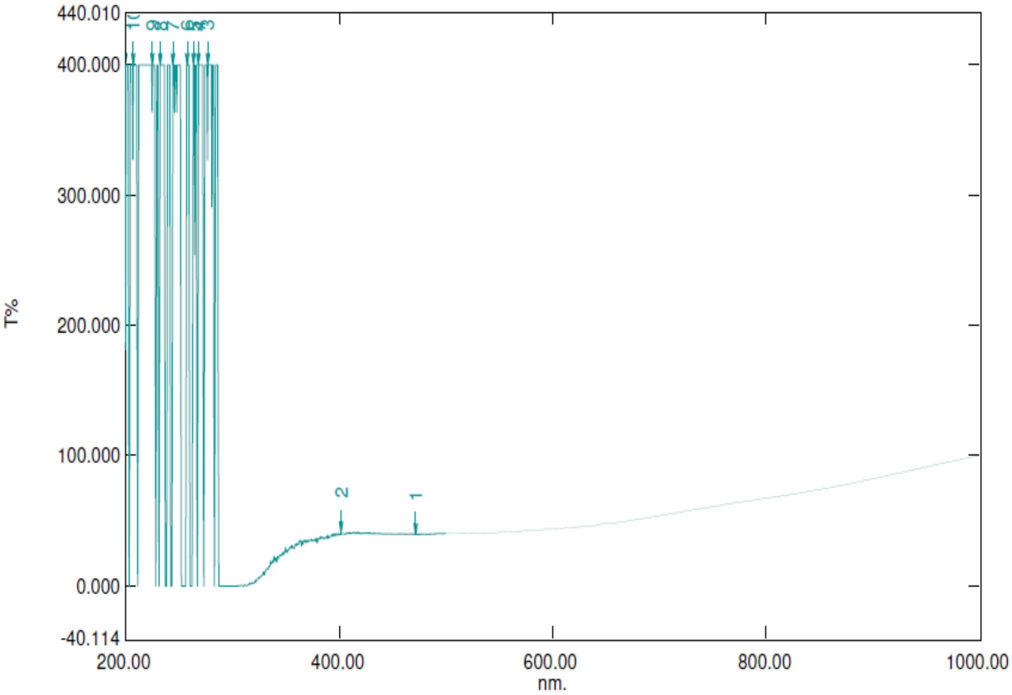


Figure 38: Transmittance of FTO /TiO_2/ZnO/Laali dye

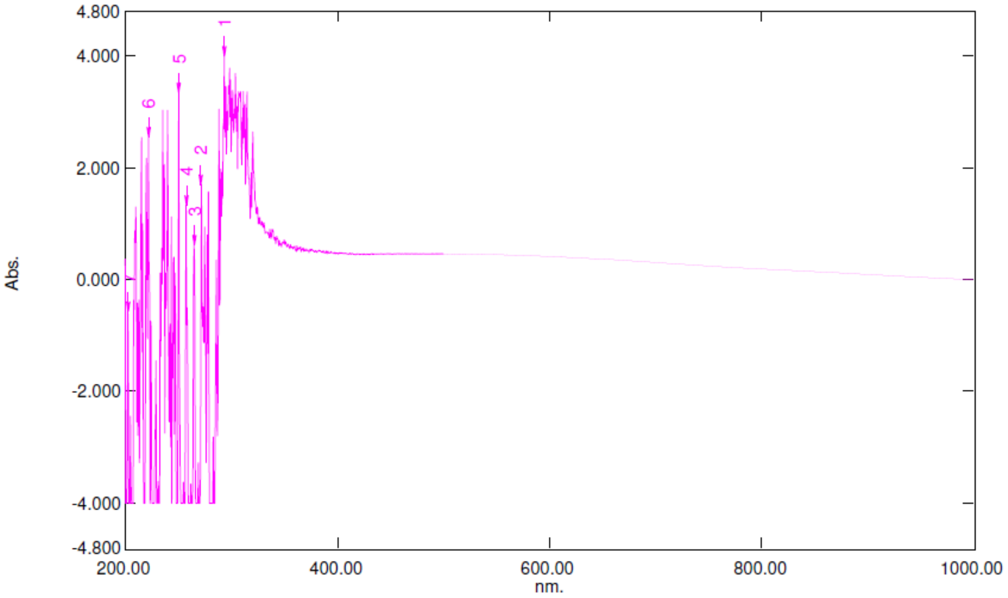


Figure 39: Absorbance of FTO/TiO$_2$/ZnO/Laali dye

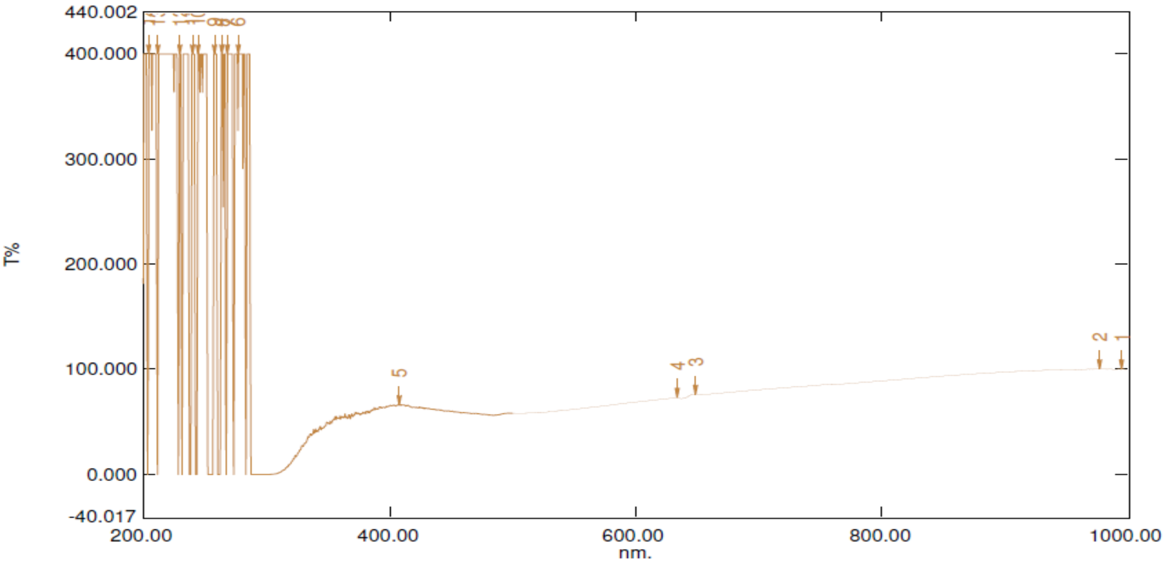


Figure 40: Transmittance of ITO/TiO$_2$/ZnO/Laali dye

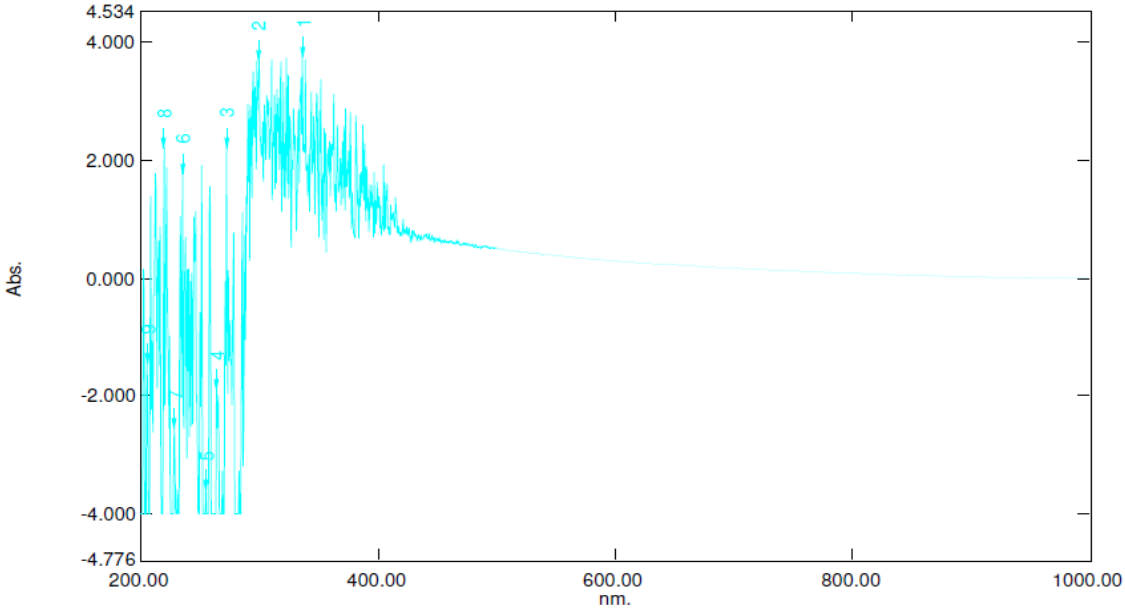


Figure 41: Absorbance of ITO /TiO$_2$/ZnO/Laali dye

IV. Discussion

In the scanning electron microscope result, Laali dye alone showed a spotted visible grain size while it was enlarged by adding ZnO which agrees with Rajaram [2] that ZnO increases the window of absorption while TiO$_2$ reduced the area of absorption. FTO approved a concise distribution of grain size area and ITO revealed the shape of Laali dye (Flowerlike in shape).

For X-ray diffraction results, Laali had no peaks on addition of ZnO the peaks increased to two and on addition of TiO$_2$ there was an improvement in the visibility of the peaks which agrees with the rutile nature. Furthermore FTO increased the number of peaks to five while ITO improved the width of the peaks.

In figure 26 of the EDS result of the Titanium dioxide there was carbon and oxygen and on introduction of ITO and the sensitizer Laali dye Phosphorous was visible which improves the electron transport in the DSSC which agrees with the findings of Prihanto [10].

In the energy dispersive spectrum, Laali dye has no atomic composition but on addition of ZnO, had four elements with carbon in abundance and Zn 12.62% while with TiO$_2$ composite system had five elements with oxygen in abundance and Ti 27.06%. Likewise with FTO composite had four elements with carbon in abundance and lastly with ITO, Had five elements with oxygen in abundance. For TiO$_2$ film carbon was the highest 51.84% with Zinc 9.90% while for ZnO film carbon was 57.08% and zinc 8.07%. The optical characterization was in the UV-Visible region which is similar to that of Mamta [5], all ZnO, TiO$_2$ anchored to the dyes, figure 28 Transmittance of TiO$_2$ shows broad edge and sharp increase at 580nm with a gradual rise. Figure 28, Transmittance of ZnO shows rise at two points 380nm and 650nmwavelength and sharp fall in absorbance edge at about 350nm wavelength and remain constant and tens to zero figure 30. Transmittance of TiO$_2$/ZnO rises at two points 390nm and 650nm wavelength figure 32. Broad absorbance edge fall at 380nm and steadily reduces figure 33. Transmittance of FTO glass rises at 350nm and is steady at two points 610nm and 620nm wavelength figure 34 while absorbance edge drops at 350nm and steadily reduces as wavelength increases figure 35. Transmittance of ITO glass rises at two points 400nm and 610nmwavelength figure 36 while absorbance edge fall at 380nm and steadily reduces to zero as wavelength increases. On the addition of FTO, ITO, ZnO and TiO$_2$ to laali dye there are visible interaction as seen in figures 37, 38, 39, 40, 41.

V. Conclusion

The result shows that ZnO enlarged the grain surface of absorption while TiO$_2$ helps to identify the shape of the image. ITO improved the number of elements contained in the dye composite system in the energy dispersive spectrum improved the peaks and arranged it in the rutile nature of titanium oxides while FTO increased peaks and gave concise distribution of grain size, with this observation, it shows that the sensitizer-laali dye anchored on these composite systems which shows their suitability materials for the fabrication of dye sensitized solar cells.

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