



An Investigation of Chemically Synthesized Barium Stannate (BaSnO_3) Thin Films Sensitized with Local Colors for Their Potential Photovoltaic Utilizations

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ABSTRACT

Three different natural dyes (Henna, Beetroot, and Purging nut) were extracted to sensitize ternary metal oxide thin film of BaSnO_3 deposited on glass substrate via electroless deposition at room temperature. Chemical baths used contained Barium Tetraoxosulphate (vi); Tin (II) Chloride Dehydrate, $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$; Sodium Hydroxide, NaOH; distilled water and calibrated per bath of organic dyes. Each deposit was set at a temperature of 90 °C for 1 h annealed at 400 °C for 1 h. The sensitized and unsensitized nano thin films remained investigated for structural, morphological, optical properties, elemental composition and electronic (chemical) structure besides existence of functional groups via X-ray diffraction (XRD), Scanning Electron Microscope (SEM), UV-VIS spectrophotometer, Energy Dispersive X-ray Fluorescence (EDXRF) and photoluminescence Fourier Transform Infrared Radiation Spectroscopy (FTIR). Polycrystalline thin films stood proved for porosity and extreme surface area for colorant loading for photosensitization in colorant sensitized solar cells (DSSCs). For both films, 1.61 eV to 1.76 eV band gaps were achieved. Unsensitized film has band gap of 1.61 eV while sensitized have greater values, hence, increased band gap and thereby violating the dye sensitization process result of E_g . Though the energy band gap of the dye-sensitized materials increased yet they remain low and this could be attributed to local state formation near the band edge. The FTIR results showed good photophysical, carboxylate and modification properties of the colorants for sunlight harvesting, anchoring and surface structure modification of films. The colorant influenced the optical characteristics of the films particularly, increasing energy band gap, E_g from an increase in absorption coefficient α , giving high absorbance A , low refractive index n , low extinction coefficient k , low reflectance R , inferring its potential applications in window coatings, solar energy devices for construction (of roofs in temperate regions), poultry houses, solar cells and DSSCs.

Keywords: Barium Stannate, Natural Dyes, Chemical Bath Deposition, Potential Applications, Solar Energy Devices

1.0 INTRODUCTION

1.1 Background of The Investigation

The forecast universal energy demands, owing to growing uncertainties of ecological contamination in the twenty first century, have inspired a big arrangement of hard work into the decrease of fossil fuel utilization resulting from investigation of hygienic, plentiful besides eco-friendly renewable energy source know-hows towards improving the value of human existence on this earth. Numerous renewable energy skills stand to be explored for assessment of their probability of addressing the rising energy need. Such springs comprise:

wind turbines, hydropower, nuclear power plants, wave and tidal power, photovoltaic (solar cells), solar thermal and so on. Amid these foundations, photovoltaic expertise, someplace sunshine is transformed into an electrical energy, has attracted substantial consideration as it changes into an exclusive besides possible result (Eze, Obodo, Ezema, & Kebede, 2021).

The entire energy technological basis that could be renewed approve one joint reverie, that is, in the direction of apprehending a kind of energy that in future will converts to treasured besides tactically imperative quality, which remains, electric energy.

As the sun offers a substantial quantity of energy for our globe, the energy it provided is roughly 10,000 times more than global demand the total energy offered by the sun is of substantial quantity for the globe and is roughly 10,000 times greater than universal energy need (Suliman, 2007) (Eze C. N., 2010) (Eze C. ..., 2010).

Metal oxide semiconductors, which are stable and environmentally friendly materials are used in photovoltaics either as photo electrode in dye-sensitized solar cells (DSSCS/DSCS) or to build metal oxide p-n junction and are equally used in some other ways in other solar energy devices (Augustine & Nnabuchi, 2016).

The utilization of multication oxides has stayed infrequently been discovered. Many ternary oxides are more stable against acid and natural dyes than their binary oxide counterpart (Augustine C. , 2007). They establish actual captivating group of resources showing a structural diversity then characteristics. They perform pertinent part in numerous ranges/parts of contemporary knowhow. There remain oxides that have metallic characteristics besides oxides that have extremely insulating/shielding behaviour. There stand oxides which navigate mutually these regimes (Eze, Onyia, & Nnabuchi, 2023).

The attention in producing numerous ternary nano films generally has come up in current ages notwithstanding noticeable hitches in producing decent ratings (Onyia & Nnabuchi, 2016).

Ternary films regularly spring decent features from the dual binary compounds (binary metal compounds) that they are made of (Onyia & Nnabuchi, 2014)

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Generally, materials including ternary metal oxides are sensitized to increase their solar energy harvesting performance. When thin films are deposited, they do well in solar energy applications. But they do better when they are sensitized because the energy band gap of the material is reduced making conduction easy and fast. Sensitizing allows the materials to absorb a wider sunlight spectrum and improve its conductivity. These materials enhance energy harvesting thereby providing solution to global energy crunches (Eze, Obodo, Ezema, & Kebede, 2021) (Eze, Obodo, Ezugwu, & Ezema, 2021).

Sensitizing really rest on the group of nanomaterial which the substance is made up of. The sensitizing procedure will differ in circumstance of metallic nanoparticles, oxides or semiconductor (e.g. chalcogenides). However, it is a usual practice that sensitizing starting materials are added at the onset of synthesis, before the nanoparticles' nucleation (NPs), in the chosen molar ratio besides this technique produce almost 100% outcome (Eze, Onyia, & Nnabuchi, 2023) (Eze, Obodo, Ezugwu, & Ezema, 2021).

Sensitizing is the careful adding of filths (adding extra constituents) inside a substance in order to change potentials of the parent substance. Such technique adds to the exceptionality of the original substance sensitized, boosting its relevance for varied areas. Sensitizing nanomaterials springs a flexible method for modifying features of the substances whilst maintaining their large surface areas. It remains expansively applied technique that modifies nanoparticles for enhancement of their electrical, optical and ecological goings-on (Das, Roy, & Devi, 2018) (Para & Reshi, 2016) (Eze, Obodo, Ezugwu, & Ezema, 2021) (Riza & Sepeal,

2019) (Shan, et al., 2004) (Ahmadi, et al., 2004). For ternary metal oxides and oxide combinations like ZnSnO_4 etc, most studies are in the context of depositing the ternary oxide thin film and characterizing for applications in photocatalysis, sensing, photovoltaics, etc.

Deliberate addition of impurities to various elements aids in altering besides moving the material on the way to needed easing features such as electronic, optical, photochemical, photo electrochemical, photocatalytic and photoexcited features. Such materials could be tailored to some specific traditions through thoughtful selecting of the dopants. Deliberation addition of impurity to nanoparticles, offers an expectation which shows that sensitized materials can get utilizations into definite area. Lately, this impurity addition to nanomaterials have lured investigative dominant support to nanotechnology for their wide practices in applied utilizations in the areas of electrical, electronics, photonics, optics, homeland security, medical sciences etc. (Eze, Obodo, Ezugwu, & Ezema, 2021).

Metal oxides thin films like organic and inorganic substances, can similarly have impurities carefully added to them (eg colors). Outcomes revealed that photovoltaic efficiency for sensitized substances is noticeably greater than unsensitized ones (Eze, Onyia, & Nnabuchi, 2023)

Ternary metal oxides of stannates and titanates such as: Zinc Stannate (ZnSnO_4); Zinc Titanate (ZnTiO_3); Strontium Titanate (SrTiO_3); Strontium Stannate (SrSnO_3); Barium Stannate (BaSnO_3) and Barium Titanate (BaTiO_3) could be promising materials for applications in solar energy devices especially in photovoltaic applications particularly when they are sensitized with some natural dyes.

Looking out for essential substitute to binary metal oxides, countless modest oxides are being fruitfully discovered like ZnO , SnO_2 , Nb_2O_5 , ZrO_5 , CeO_2 , Al_2O_3 , In_2O_3 and Fe_2O_3 . These oxides have been studied individually before in the system of metal oxide-metal oxide composite, sensitized metal oxide or core-shell structure which have been done broadly. We have comparatively limited researches of complex oxides remaining. There are rising awareness in multifaceted oxides which arrives in form of features of binary oxides, whilst glowing recognized in more than a few areas, appear to limit the functioning of novel utilizations (Karst, 2011) (Zumeta, 2016).

Ternary metal oxides were sought for because they are better and more versatile in terms of synthesis and applications. When they are sensitized, they harvest sunlight energy better and yield enhanced solar energy applications in some solar energy devices. Sensitizing allows the materials to absorb a wider sunlight spectrum and improve its conductivity. This remains predominantly fascinating in energy utilizations anywhere a request is made for expressly considered semiconductors for improved contest of the characteristics of developing resources.

Stannates with cubic perovskite structure (MSnO_3 , $\text{M} = \text{Ca}, \text{Sr}, \text{Ba}$) have exciting high-tech utilizations, such as thermally unchanging capacitors, lithium batteries, catalysts, pigments, sensors etc.

Barium stannate (BaSnO_3) with a standard band gap of about 3.0 eV or more remains a supreme auspicious developing resource that can be used as a transparent electrode. However, our research results depict 1.61 to 1.76 eV. Up till now, its synthesis stands to be recognized as being seriously difficult to deposit.

Barium Stannate remains extremely water insoluble crystal-like Barium basis that can be used in well-matched advanced (elementary) PH settings. It remains directly accessible in ultra-high purity besides high purity configurations that progress mutually photosensitive value besides helpfulness by way of systematic values. It is an oxide of barium and tin with the chemical formula: BaSnO_3 and empirical formula is O_3BaSn . It appears as powder or particles. Density is 7.6 g/cm^3 and molecular weight of 304.03. It has high mobility, transparent in the visible range, good thermal stability, versatile applications and good electromagnetic shielding due to its low sheet resistance etc. It remains extensive band gap semiconductor with investigational principles of that gap changing on or after 2.95 - 4.05 eV

and with a perovskite crystal structure) It keeps huge carrier mobilities (μ) for n-type conduction at room temperature at comparatively great carrier densities (n) eg $\mu \leq 320 \text{ cm}^2 \text{ V}^{-1} \text{ S}^{-1}$ at $n \sim 8 \times 10^{19} \text{ cm}^{-3}$ (Eze, Onyia, & Nnabuchi, 2023)

BaSnO₃ can be used as a gas sensor, transparent conducting film, etc

Choosing a particular synthesis technique requires diversity of contemplation and principles like thin film utilization, substance uniqueness, in addition to procedure know-how. Is very clear that there exists no common rule for selecting the most excellent synthesis process. Nevertheless, diverse measures and synthesis expertise with materials' and substrates' type, particularized utilization, expenditure, as well as demanded efficiency makes provision for the selection of a suitable method. These processes can be chemical or physical (Augustine & Nnabuchi, 2016), Zumeta, I.A. (2016). TiO₂ films got by microwave-activated chemical-bath synthesis employed for progressing TiO₂ conducting glass contact. *Solar Energy Materials and Solar Cells*, 93 (10): 1431—1439.

Many researchers have deposited BaSnO₃ in a variety of ways. (Jibi, et al., 2016) deposited BaSnO₃ via Solid State Ceramic Method. (Jingyeong, Youngkyoung, MacManus-Driscoll, & Shinbuhm, 2023), grew La-sensitized Barium Stannate (BLSO) films on industrially practical optoelectronic MgAl₂O₄ substances for TC-EMS. (Merve, Sibel, Alper, & Serdar, 2021) synthesized BaSnO₃ using Flame Spray Pyrolysis, other methods are: Sol-Gel method, Hydrothermal Synthesis, wet chemical route (Rakesh, Sharmila, & Pamendra, 2018), Auto-Ignited Combustion Technique, Excimer Laser-assisted Metal Organic Decomposition (ELAMOD) (Panel, Junichi, Iwao, Nishio-Hamane, & Tetsuo, 2020) etc.

The deposition of these our thin films is through chemical method via Solution Growth (SG)/Chemical Bath Deposition (CBD) routes. Deposition methods will help to improve the quality of film got (Ezema, 2004).

The electroless technique has shown that for thin films synthesized via autocatalytic growth method, the procedure of nucleation as well as film synthesis is predominantly owing to a joint interaction of added methods that comprises: simple ion cluster mechanism, simple hydroxide cluster mechanism, complex ion-by-ion decomposition mechanism and the complex-cluster decomposition mechanism (Ezema, 2004). The chemical bath deposition procedure typically hires a meticulous chemical response for attaining thin film synthesis via precipitation of the anticipated composite.

The solution growth technique has been termed a solution growth technique or electroless deposition or autocatalytic deposition. It stands a process for synthesizing thin films as well as nanomaterials first described in 1969. This could be used for great expanse group processing or incessant synthesis. This remains the cheapest, suitable and modest method which is readily reproducible. The method stays applied to synthesis of conducting besides non-conducting sheets from solution by electrochemical process when there is nonappearance of an external field (Quijada, Para, Reshi, & Loesch, 1998) (Eze, Onyia, & Nnabuchi, 2023). It makes use of the idea that films could stay synthesized on the substrate if metallic or non-metallic thru plunging them inside appropriate solution steam bath holding metal salts. The metal ions besides non-metallic ions that appear inside the growth mixture react with one another to produce unbiased mixtures. Such process stays founded on a skilful precipitation of the wanted composite from a reaction mixture.

In Chemical Bath Deposition method, growing thin films via controlled ion-by-ion reaction spontaneous precipitation, essentially should remain circumvented. Slowing down the reaction has to be via using appropriate complexing agent or ligand in a appropriate intermediate showed in the pH of the reaction mixture. Such remains This is the foundation of chemical bath deposition process.

Investigational arrangement of the CBD remains modest, low-priced as well as expedient. The glass

slide substrate is inserted straight up then at the fitting midpoint of the solution immersion container. The bath is curtained from dust besides additional ecological filths using artificial foam protection.

The reaction mixture is regulated by the substance to be synthesized. A big benefit of this method remains its ability to synthesize unvarying films on unevenly designed substrate besides also at facts unreachable via other methods.

Looking at this investigation, autocatalytic process has been employed to grow a uniform natural dye-sensitized thin film material of BaSnO_3 . All the plants that were used for the extraction of natural dyes in this study were got from within Nsukka area of Enugu State in Nigeria and they are; Henna (Laali/Lalle), *Jatropha curcas* (Obobolochi) and Beetroot.

- i. Henna or Hina (common name); Leaves of *Lawsonia inermis* (Botanical name); Laali (Ibo); Lalle (Hausa).
- ii. Beetroot (common name); Roots or fruits of *Beta vulgaris* (Botanical name).
- iii. Purging nut (common name) *Jatropha curcas* (Botanical name); Obobolochi (Ibo name); leaves of the plant.

1.2 Problem Statement

Fossil fuel is not a lasting solution to global energy demand because it will deplete. They are ecologically unfriendly, expensive and not readily available. Going by this, this work addresses the following:

- i. The practice of accessible, low-priced then naturally benign resources such as home-grown colors for the capture of plentiful astral energy.
- ii. The use of stannate, that is tunable, inexpensive then hopeful besides functional in DSSCs, etc providing large surface area for solar absorption thereby offering result to worldwide energy shortfalls.
- iii. Development of cheap, hygienic also renewable energy know-hows and the utilization of environmental energy via solar cells, photo-electrochemical cells, dye-sensitized solar cells (DSSCs) besides perovskite solar cells (PSCs) which offers energy request solution in developing countries like Nigeria that find it difficult to utilize this sunlight energy due to the cost of materials and technological know-how in processes and applications.

1.3 Research objectives

The growth of ternary oxide-based thin films of Stannates (BaSnO_3) sensitized with diverse local dyes via chemical routes

Characterization, investigation then evaluation of the thin films sensitized with dyes.

Assessment of their appropriateness then possible utilizations in solar energy devices.

1.4 Contributions

Here is the foremost stint where CBD technique is employed to grow thin films of Stannates (BaSnO_3) sensitized with diverse local dyes, albeit, highly difficult in synthesizing the said thin films.

The innovation is in the practice of employing low-priced resources as well as simple technique to resolve universal energy crisis.

Investigational study which generated effective then relevant information/outcome meant for technical applications.

1.5 Related Work

J. Jibi, *et al.*, carried out research on the Deposition, Structural and Morphological Characteristics of BaSnO_3 Nano powder Processed by Solid State Ceramic Procedure. They got a stoichiometric amount of BaCO_3 and SnO_2 powders and mixed using acetone in an agate motor for 6 hours. They heated the similar/standardized solution in an alumina crucible at 1000°C leaving the mixture at same temperature for 8 hours. To fabricate pellets,

they added Polyvinyl alcohol to the calcined powder. They applied a load of 5 ton to get a circular disc shaped pellets which was further crushed and sintered in a platinum crucible at 1250 °C for 6 hours. They did characterization for XRD, SEM, TEM, EDX and UV-Vis. Their results showed a cubic structure of nanocrystalline BaSnO_3 with a porous surphase morphology, high optical reflectance and band gap energy of 3.1 eV. (Jibi, et al., 2016).

J. Jingyeong, *et al.*, grew La-sensitized Barium Stannate (BSO) films on industrially practical optoelectronic MgAl_2O_4 substances for TC-EMS. They showed that metallic sheet resistances at the single-crystal level can be attained thru dipping an MGO template level can be attained via dipping an MGO template layer amid the film besides the substrate. Their result showed a great visible transmittance film and X-band shielding efficiency (SE). Their XRD and STEM results showed that their templated-epitaxy-ideology brought a serious improvement of film crystallinity. (Jingyeong, Youngkyoung, MacManus-Driscoll, & Shinbuhm, 2023) .

Z.O. Merve, et al., studied the Characterization and Optical Gas Sensing Properties of BaSnO_3 Deposited through Novel Procedure: One-Step Flame Spray Pyrolysis using $\text{Ba}(\text{CH}_3\text{COO})_2$ and

$\text{SnCl}_4 \cdot 5\text{H}_2\text{O}$ salts as starting materials. Initially, they dissolved ethylene glycol and citric acid as a 4:1 ratio at 70°C, in addition to 0.2 M $\text{Ba}(\text{CH}_3\text{COO})_2$ then $\text{SnCl}_4 \cdot 5\text{H}_2\text{O}$ stayed additionally put to this mixture. The composition remained turned till a crystal-clear solution was attained. They divided the last mixture into dual identical quantities. For the FSP process, the occasioned mixture stood given to flame spray pyrolysis apparatus (Tethis Nps10) by means of the glass syringe to get BSO particles. To study the varying in phase structure of BSO particles, every powder was annealed at dissimilar temperatures like 600°C, 800°C, 1000°C, and 1200°C, correspondingly. Trying to enable the explanation of such outcomes, BaSnO_3 particles deposited by FSP remained encoded as BSO_FSP. They deposited ceramic powders and studied their structural, morphological then optical characteristics. Their result showed a barium crystal structure with obtained gap of 3.14 eV. They were able to measure the intensity then decay time-based gas sensing characteristics of BaSnO_3 entrenched in ethyl cellulose thin films when visible to the vapors of ethanol, acetone and ammonia. (Merve, Sibel, Alper, & Serdar, 2021).

K. Rakesh, et al., used wet chemical technique to deposit barium stannate powders. The researchers

BaCl_2 (Aldrich, 99.5%), $\text{SnCl}_4 \cdot 5\text{H}_2\text{O}$ (Sigma, 99.5%), NaOH (BDH, 99%) precursors by putting together 20.823 g of anhydrous barium chloride (BaCl_2) then 35.051 g of stannic chloride (SnCl_4) in 100 ml deionized doubled distilled water. A 4 g sodium hydroxide was put in the mixture. Such mixture stood aged for 24 h, a white precipitate was attained in the lower surface of solution. After filtering, they repeatedly washed the product with purified water to remove Cl^- ions. The white powder stood desiccated at 80°C for 24 h by means of a muffle furnace. Lastly, the dehydrated barium stannate powders were annealed at 800°C in ambient setting. The researchers studied the powder structurally by means of X-ray diffraction figures that stood measured on X-ray diffractometer (Rigaku Smart Lab, Japan). For the researchers to do impedance measurements, the well annealed powder stood pressed as cylindrical pellets (diameter 10 - 12 mm and 1 - 2 mm thickness) when a limited drips of poly vinyl alcohol (PVA) as binder must have been added. They also annealed the ready-made powder at 800°C temperature in furnace for 5 h. After firing the samples were furnace cooled. They eventually coated the pellets using conductive silver paint then desiccated at 500 °C for 2 h earlier carrying out dielectric then impedance measurements. The proportion of porosity stayed found by means of measured investigational density (Archimedes principle besides via finding the weight, area and thickness of the pellet) and by deducing the theoretical density from XRD data. The XRD investigations were made at room temperature in the Bragg angle range $20^\circ \leq 2\theta \leq 80^\circ$ with $\text{CuK}\alpha$ radiation ($\lambda = 1.5418 \text{ \AA}$) using $\text{CuK}\beta$ filter. The complex impedance measurements over temperature range from 35°C - 300°C stood accomplished by means of a computer-controlled impedance analyzer (HIOKI LCR Hi-TESTER MODEL 3532-

50) in the frequency range 1 kHz to 1 MHz. Scanning electron microscopy (SEM) is used for the analysis of surface microstructure along with EDAX spectra. DC conductivity was measured by (Keithley) 6514 system electrometers. Room temperature FT-IR and Raman spectra were measured using Shimadzu 7800 FTIR and Technos micro-Raman spectrometer respectively. Optical band gap predictable from absorption data is ≈ 2.94 eV, very near to the stated value. Composition prepared via the noble wet chemical route takes high electrical conduction then suitable for gas sensing utilizations.

2.0 MATERIALS AND METHODS

2.1 Materials

We collected those greeneries of *Lawsonia inermis* and *Jatropha curcas* from their natural stands of those species in Nsukka, Enugu State of Nigeria. Equally, root/fruits of *Beta vulgaris* was purchased from a local market in same area.

Barium Tetraoxosulphate (vi)/Barium Sulphate BaSO_4 (Sigma Aldrich, 99.0 %), Tin (II) Chloride Dehydrate, $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$ (Sigma Aldrich, 98 %), Sodium Hydroxide (NaOH) (Sigma Aldrich, 95 %) and distilled water are of analytical grade, were undiluted and as purchased and used for the growth of our thin films without further purification. These are precursors sources of Barium and Tin and reducing agent respectively used for the dye-sensitized and unsensitized films.

2.2 Dye extractions

Simple techniques were employed in the extraction of natural/local dyes used in the sensitizing of the thin films of BaSnO_3 synthesized. 12.5 g of the leaves of *Lawsonia inermis* was washed and shade dried and ground into powder. 12 g of *Beta vulgaris* was washed and peeled then rewashed and ground into pasty mass. In both cases, mortar and pestle were used in grinding. Then 12.5 g of leaves of *Jatropha curcas* was washed and boiled in 30 ml of water for 10 mins using pot and heater. Each was soaked in 5 ml of water and ethanol respectively for 2 hrs and filtered using filter paper and funnel in that order. The dye extracts collected were stored in cleansed, labelled bottles and stored in a cool cupboard at room temperature.

2.3 Synthesis of BaSnO_3

This remains one of the most hopeful developing resources to be used transparent electrode. But, up till now, the deposition remained established to be extremely unreproducible.

The synthesis procedure involving preparation of solution and deposition was utilized to obtain an

acceptable result. At optimized conditions, A 0.16 M 4 g of BaSO_4 was measured and dissolved in 50 ml of water in a beaker. A 0.1 M 1g $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$ was measured and put into another beaker and each stirred using magnetic stirrer for 10 mins. A 30 ml of BaSO_4 was put in a beaker and 20ml $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$ was put in the same beaker and stirred for 10 ml. A 5 ml of NaOH calculated at 1.0 M 0.8 g was added into this solution and stirred for 10 mins.

The first bath labelled AD (31) is the as-deposited. Three other experimental baths had the addition of three drops per bath of organic dyes of Beetroot (BR), *Jatropha curcas* (OO) and Henna dye (LL), and labelled 32, 33 and 34 respectively. A glass substrate inserted into this mixture for deposition measuring $2.24 \times 2.24 \times 0.75$ was earlier cleansed in concentrated hydrochloric acid, cleaned in cleansing agent solutions, dipped inside purified water in addition allowed to dry in a setting devoid of dust. It was inserted into this mixture through a synthetic foam for deposition in oven at 90°C aimed at 1 h and thereafter annealed at 400°C for 1 h. For comparison between the unsensitized and different dye-sensitized films, separate beakers were used and labelled maintaining the same conditions.

2.4 Characterizations

The as deposited and dye sensitized thin films were examined for crystalline structure and phase properties using an advanced X-ray diffractometer (XRD) using the energetic monochromatized $CuK\alpha$ radiation source ($\lambda = 1.5406 \text{ \AA}$) which scanned films uninterruptedly as 2θ varied from 10 to 80° at a step size of 0.02° besides at a phase period of 0.2 s . The SEM was carried out using PRO:X:800-07334 Phenom World, MVE0570775 operated at 10 KV . The UV-VIS spectrophotometer was employed in the investigation of optical absorptions of the products. The significance differences between the sensitized and unsensitized are observed and some parameters got from our UV-VIS spectrophotometer highlighted most of the possible uses of this work in astral energy devices. The constituents of those elements were studied employing EDXRF using ARL QUANT'X EDXRF Analyzer, SRM 2710 at 10 mins run time. The Fourier Transform Infra-Red Spectrometer (FTIR) Cary 630 was hired to examine chemical/electronic structure and occurrence of efficient clusters. We considered the transmittance of energy compared to wave number in a Mid-infrared spectrum (mid-IR spectrum) of the diverse dyes.

3.0 RESULTS AND DISCUSSION

3.1 The structural study

Figure 2 demonstrations the room temperature X-ray diffraction outline of $BaSnO_3$ thin films arranged via electroless procedure. The XRD shape illustrates structural peaks only without any impurity (AD/31) and others are structural peaks of dye-sensitized films.

The XRD shape displayed asymmetrical polycrystalline thin films having dissimilar high and low peaks. Intense and narrow peaks remained visible at (2θ) angles of 31.35 , 44.14 , 54.51 , 64.25 , and 72.38 , degrees and these correspond to (hkl) orientation of 110 , 200 , 211 , 220 , and 013 respectively.

These diffraction peaks confirm the deposition of $BaSnO_3$ nanoparticles prepared and deposited using CBD techniques. Its crystallographic parameter in relation to the crystal system is cubic structure. The development of single-phase substance remains established by way of not at all extra XRD outlines owing to starting materials of oxides or minor secondary phases stand observable inside the discovery border. The enhanced diffraction peaks occurred at various 2θ values confirms the sensitizing of $BaSnO_3$ with different dyes which enhances light absorption and makes electron mobility faster. Such investigational XRD outline settles with the Joint Committee on Powder Diffraction Standards (JCPDS) card number: 74-1300 ($BaSnO_3$). The XRD spectrum is equally greatly in line with what was earlier reported by (Rakesh, Sharmila, & Pamendra, 2018) (Wu, 2012).

The average crystallite size found by means of Debye-Scherrer technique is 23.55 nm . Small crystallite size could stay an important motive.

The Debye-Scherrer equation (Bown, Pardo, & Mombru, 2019) is specified in equation 1 thus:

$$D = K\lambda/\beta\cos\theta \quad (1)$$

$$\text{Lattice spacing, } d(\text{nm}): d = \lambda/(2\sin\theta) \quad (2)$$

$$\text{Dislocation density, } \delta: \delta = \Delta/(D^2) \quad (3)$$

$$\beta(2\theta) = 4\epsilon\sin\theta/\cos\theta \quad (4)$$

$$\text{Lattice Strain, } \epsilon: \epsilon = \beta/2\sin\theta \quad (5)$$

$$d = \lambda/2\sin\theta \quad (6)$$

we state that, β is known as the peak's full width at half maximum (FWHM) intensity (ie half of peaks width), K shows a constant identified as the shape factor roughly equivalent to 0.94 , λ has a value of (0.1542 nm) or $1.5406 \text{ \AA}$ and stands for the wavelength of the one-color sunlit needed in irradiating that material. D means crystallite magnitude then θ means Bragg's angle.

Gaussian fit algorithm was used to run an nonlinear fit (NLF) on the peaks, this is attributed to the fact that it gives less standard deviation values after fitting than other fit algorithms.

The dislocation density was estimated using equation 3. The peak broadening due to microstrain varies as shown in equation 4. The estimation of the lattice strain, ϵ , was done as seen in equation 5. The inter planer distance, d , that remains the vertical spacing between the parallel atomic planes in a crystal, remained got as seen in equation 6.

Table 1 shows a summary of the lattice parameters for the film such as the lattice constant (a), lattice spacing (inter planer spacing) (d), Lattice Strain (ϵ), FWHM (β), crystalline size (D) then dislocation density (δ).

The average crystallite size of the BaSnO₃ film gave a value of 23.55 nm with an average lattice strain of 0.2724. The occurrence of lattice strain remained distinguished owing to the interstitial filths found inside sample besides the non-uniform distribution of d -spaces in the crystallographic plane (Speakman, 2015).

The average crystallite size and lattice strain of the BaSnO₃ at deposition temperature of 90 °C and annealing temperature of 400 °C is 23.55 nm and 0.2724 respectively.

The presence of lattice strain has been noted to be due to the interstitial impurities in the sample and the non-uniform distribution of d -spaces in the crystallographic plane (Speakman, 2015).

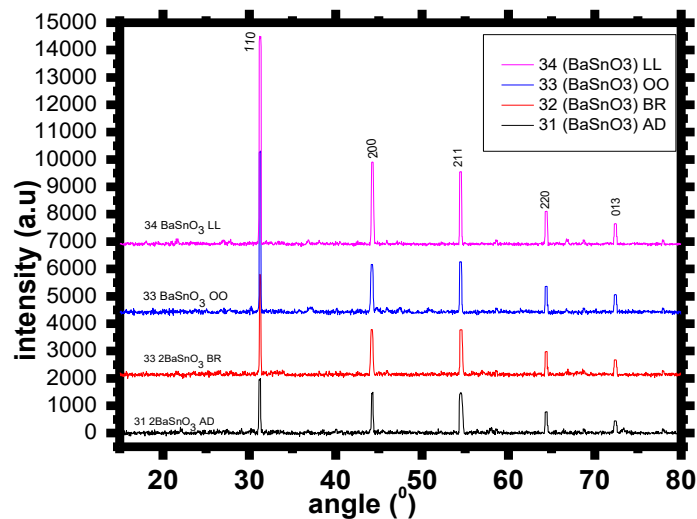


Figure 1: The XRD shape of CBD deposited BaSnO₃ of AD films and films sensitized with different dyes

The Table 1 presents the Lattice parameters. The table for d -values matches well with the d -table obtained by (Rakesh, Sharmila, & Pamendra, 2018).

Table 1: The quantities obtained after the XRD outline of the BaSnO₃ sensitized with different dyes

Angle (2 θ)	θ in rad	FWHM (β)	FWHM β (in rad)	Crystallite Size, D (nm)	Lattice spacing, d (nm)	Dislocation density, δ	Lattice Strain, ϵ
31.21233	0.272379	0.46892	0.008184	17.59016	2.863319	3.23E+13	0.419696
44.23878	0.386056	0.46724	0.008155	18.35337	2.045744	2.97E+13	0.287389
54.51213	0.475708	0.29913	0.005221	29.87505	1.681995	1.12E+13	0.145161
64.26217	0.560793	0.43708	0.007628	21.46328	1.448319	2.17E+13	0.173982
22.38161	0.195316	0.26565	0.004636	30.48472	3.969045	1.08E+13	0.33569
Average				23.55332	2.401684	1.8E+15	0.272384

3.2 Surface Morphology

Surface Morphological Studies of Barium Stannate (BaSnO₃) Nanoparticles

The figures 2 – 5 shows a SEM micrograph of the Barium Stannate, BaSnO₃ sample. The SEM image shows micrograph of cubic structured BaSnO₃ sample sensitized with BR, OO and LL dyes and As- Deposited samples. The samples showed nanograins/pins appearance. Sample AD showed ridges of nanograins/nanopins of different sizes and shapes with boundaries and patches of holes of different diameters. Sample BR showed fine grains with very few circles of grains/pins surrounded by empty spaces. Sample OO have nanograins of pebble-like/sponge-like structure presenting flower-like clusters aligned in the same direction. BR and OO exhibited a more increased sizes of the nanopins structures with BR forming spongy clusters. Sample LL have larger sized nanograins/pins of different sizes of about 0.00855 nm (0.855) μm with big agglomeration at a location. It has larger pores than others equally exhibiting surface sample modification due to the dye content. sample LL formed mesoporous surface/flower-like clusters with minor cracks. The ccd (LL) sample is shown to have better photophysical property than others. is important indication of the presence of a property needed in a dye-sensitized sample for dye loading and photosensitization in DSSCs.

BR is in line with what was reported by (Rakesh, Sharmila, & Pamendra, 2018).

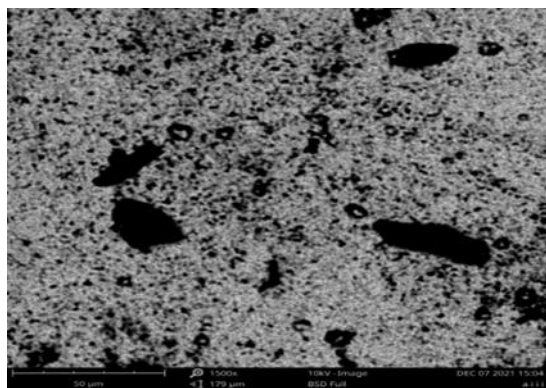


Figure 2: The SEM Micrograph of Model 31 of BaSnO₃ As- Deposited (AD)

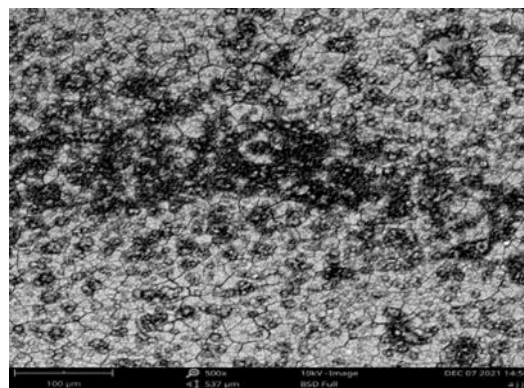


Figure 3: The SEM Micrograph of Sample 32 of BaSnO₃ Sensitized with Dye3 (BR)

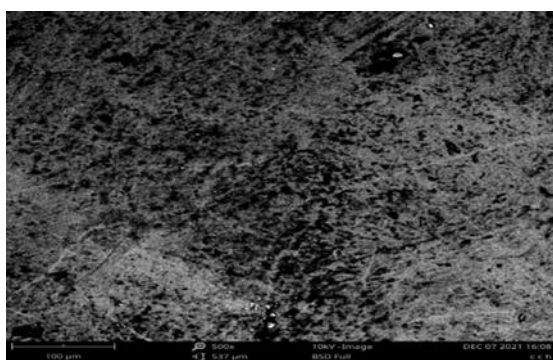


Figure 4: The SEM Micrograph of Model 33 of BaSnO₃ Sensitized with Dye2 (OO)

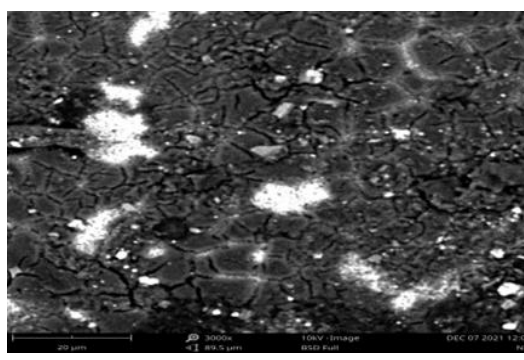


Figure 5: The SEM Micrograph of Sample 34 of BaSnO₃ Sensitized with Dye1 (LL)

3.3 Optical Analysis/Studies of Barium Stannate (BaSnO₃) Thin Films

Figure 6 displays the plot of absorbance versus wavelength in the wavelength range 229.99 to 1099.78 nm and 300 nm to 1100 nm. The four samples have good, strong, high and steady absorbance within the visible region of the electromagnetic spectrum from 370 nm to 722 nm. From the point of absorption band edge to 800 nm, the absorbance is equally reduced due to the dye content. The As- Deposited, AD BaSnO₃ film has absorbance band edge of 588.67 nm

and absorbance of 41 %. The BaSnO₃ film sensitized with Henna dye (LL) exhibited absorbance in the visible region with absorbance band edge of 488.42 nm and absorbance of 26 %. Sample OO has absorbance band edge of 572.14 nm and absorbance of about 38 %. Sample BR has absorbance band edge of about 469.60 nm and absorbance of about 33 % within the visible region. The dye sensitized samples have lower absorbance than the AD sample. There is shift towards higher energies resulting to higher/increased energy band gaps. The sample sensitized with Henna has lower absorbance than other samples sensitized with dyes. Here influence of the plant dyes reduced absorbance significantly independent of the dye content.

Figure 7 shows the spectral distribution of reflectance of BaSnO₃ for unsensitized and sensitized samples. The reflectance was calculated using the relation shown in equation 7 thus:

$$R = 1 - A - T \quad (7)$$

All the samples have low reflectance. For samples AD and OO, reflectance increased with increase in wavelength from 500 up to 585.42 nm and then decreased with increase in wavelength from up to 1049.60 nm and finally, reflectance increases with increase in wavelength within a very small region.

From the spectrum, the average optical reflectance of all samples was about (0.20) 20 %.

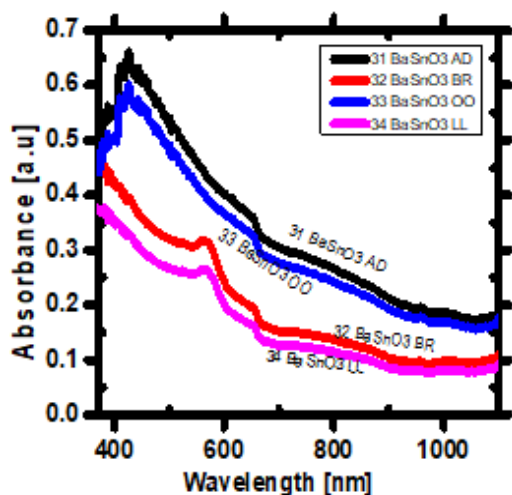


Figure 6: Absorbance Graph of BaSnO₃ Thin Film Sensitized with Dyes and as-deposited

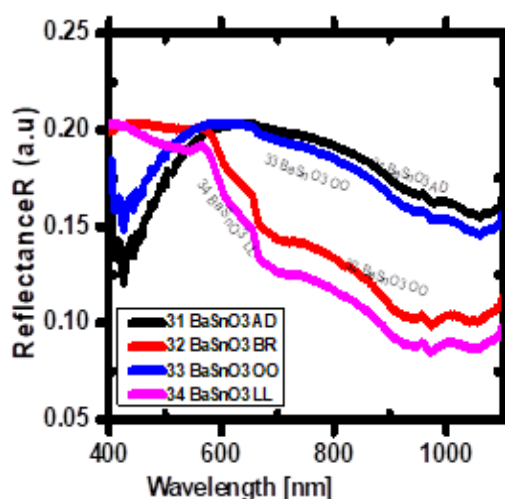


Figure 7: Reflectance Graph of BaSnO₃ Thin Film Sensitized with Dyes and as-deposited

Figure 8 shows the transmittance spectrum of both the unsensitized, AD and the samples sensitized with dyes. The transmittance increases with sensitizing. The transmittance of the unsensitized is about 22 % while the sensitized are 24 % for sample sensitized with dye OO, 36 % for sample sensitized with dye BR and 42 % for sample sensitized with dye LL. The low transmittance may be attributed to the fairly high crystallite size of the film as observed from the XRD analysis on table 1. It is well known that when the crystallite size is fairly large, the smoothness of the film decreases which cause detraction in the optical transmittance. Lower transmittance specifies higher imperfection density besides poor electrical characteristics for the sensitized. This is due to absorption of light in the longer wavelength area of say more than 500 nm is mostly due to crystalline imperfections like grain boundaries besides dislocation (Jutika, Dhanapati, & Ashim, 2011).

Such sensitized samples have greater transmittance than that unsensitized one. Equation 7 can be used to calculate the transmittance. Sample LL has a comparatively higher transmittance than other dye-sensitized counterparts. All the dyed samples have applications in window coating of poultry houses for warmth and to reduce the use of lantern and stoves.

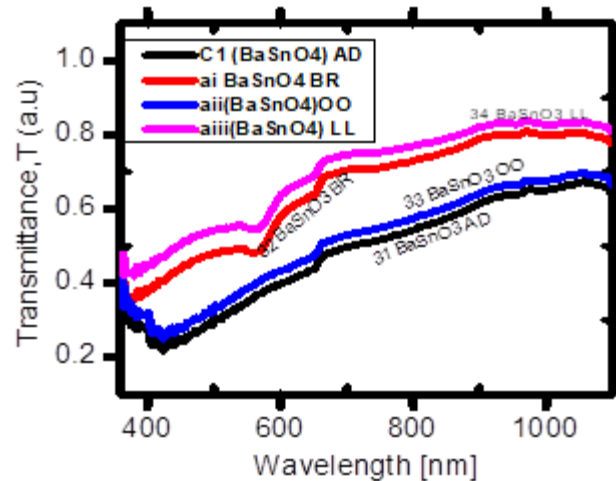


Figure 8: Transmittance Graph of BaSnO3 Thin Film Sensitized with Dyes

Figure 10 depicts the estimated optical energy band gap E_g of the BaSnO3 for both the As-Deposited, AD and other samples sensitized with extracted natural dyes from Henna leaves, LL; Purging nut, OO; and Beetroot, BR. In doing so, we used the absorbance data in the wavelength range covering the visible region of the electromagnetic spectrum to estimate the absorption coefficient α . A plot of $(\alpha h\nu)^2$ vs. $h\nu$ was done using Tauc's model and also Davis and Mott model stated in equation 8 thus:

$$(\alpha h\nu)^2 = A (h\nu - E_g) \quad (8)$$

Figure 9 shows the extrapolations of the graphs showing that sensitizing BaSnO3 films has effect on the optical band gap energy of the samples. The optical band gap energy increased from AD (31) = 1.61 eV to 33 (OO) = 1.67 eV, 34 (LL) = 1.99 eV and 32 (BR) = 1.76 eV. There is increase in E_g for the samples sensitized with natural dye. This, however, violates the dye sensitization process result for energy band gap, E_g . All the samples have low energy band gap thereby finding their applications in window coatings.

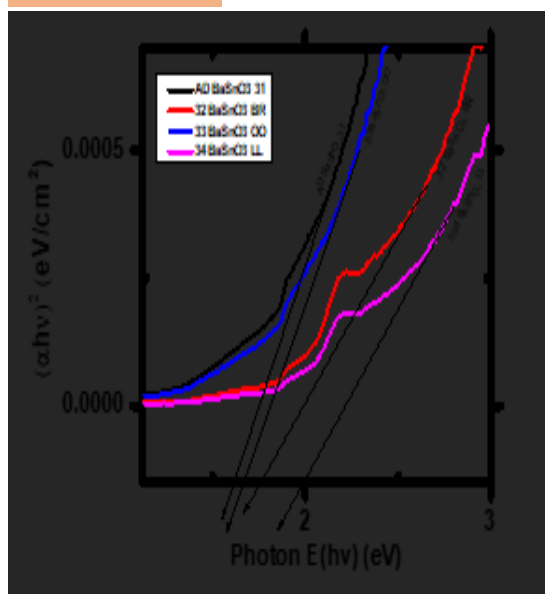
The low energy band gaps we got, in this work, are albeit lower than the reported results of (Rakesh, Sharmila, & Pamendra, 2018). This smaller value of E_g according to our research in comparison to the standard value of 3.1 (generally, 2.9-4.2) eV could be attributed to local state formation near the band edge

Figure 10 shows the extinction coefficient k of BaSnO3 samples for both As- Deposited, AD and

other samples sensitized with three natural dyes extracted from Henna, LL; Purging nut, OO and Beetroot, BR. The variation of the extinction coefficient k of such samples is found from equation 9 stated (Karst, 2011) thus:

$$k = \alpha\lambda/4\pi \quad (9)$$

The values of k which decrease by varying wavelength of the incident photon were obtained. For the AD sample, k decreased from 0.49 – 0.42 by varying the wavelength from 425.45 – 601.40 nm; for the sample sensitized with OO, k decreases from 0.45 – 0.39 by varying the wavelength from 434.00 – 594.83 nm; for the model sensitized with Henna, LL, k cuts from 0.25 – 0.23 via varying the wavelength from 376.12 – 481.36 nm and for the sample sensitized with Beetroot, BR, k decreased from 0.29 – 0.28 with a corresponding increase in wavelength from 380.39 – 496 45 nm. The AD sample has higher k followed by OO, BR and LL samples. The dyed samples with a comparatively lower k and a small energy band gap E_g , and low n , they all have good qualities to be applied in window coatings.



AD (31) = 1.61 (eV), (34) LL = 1.99 (eV),
(33) OO = 1.67 (eV), and (32) BR = 1.76 (eV).
Figure 9: Energy Band Gap Graph of
BaSnO₃ Thin Films Sensitized with Dyes

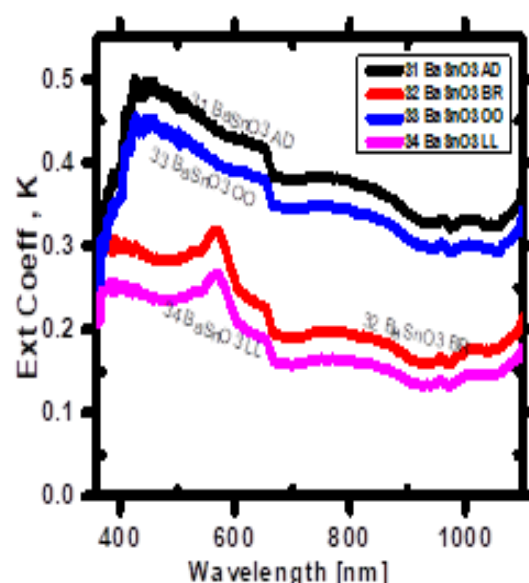


Figure 10: Extinction Coefficient Graph of
BaSnO₃ Thin Films Sensitized with Dyes

3.4 Composition Study

The EDS results show spectrum of sample 31 (AD), 32 (BR), 33 (OO) and 34 (LL) of BaSnO₃ nanostructures establishing that the sensitized and as-deposited thin films have their different

elements properly represented as shown in figures 11, 12, 13, and 14. Other elements found in the compound are from substrates(glass) and dyes as beetroot, henna and some other dyes is known to contain potassium, sodium, phosphorous, calcium, magnesium, copper, iron, zinc, nitrogen and manganese (Hathan & Singh, 2014), (Sakuntala, Raju, & Jaleeli, 2019) (Boubaya, Marzougui, Yahia, & Ferchichi, 2011). However, Barium was detected in low quantity in accordance with the earlier stated fact of its characteristic irreproducibility and strenuous deposition attributes particularly via CBD route.

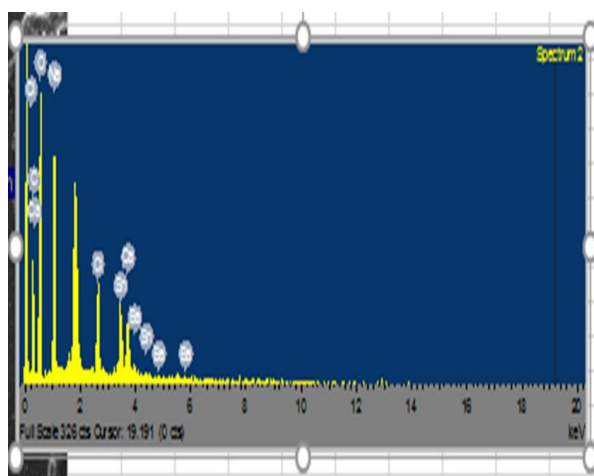


Figure 11: The EDS Spectrum of Sample 31
of BaSnO₄ Nanoparticles As- Deposited (AD)

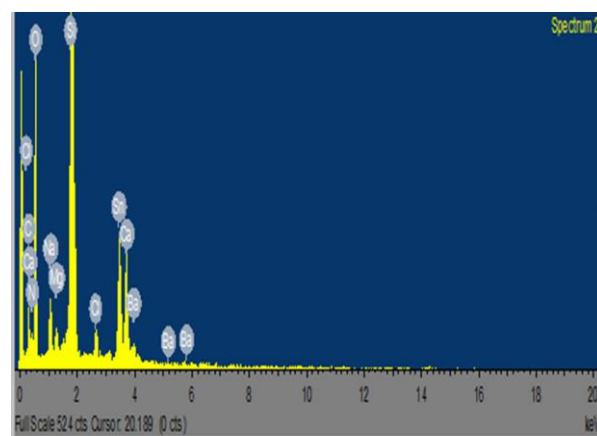


Figure 12: The EDS Spectrum of Sample 32
of BaSnO₃ Nanoparticles Sensitized with Dye
D3 (BR)

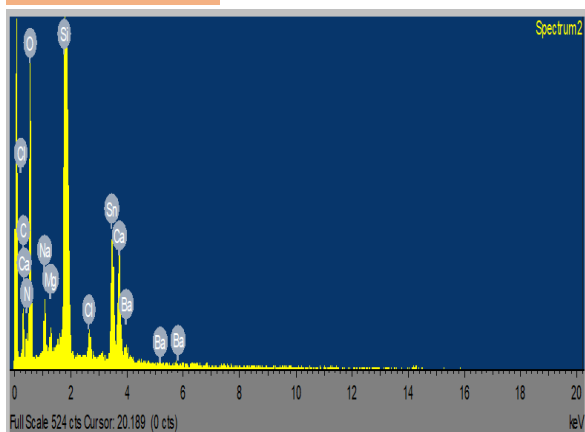


Figure 13: The EDS Spectrum of Sample 33 of BaSnO₃ Nanoparticles Sensitized with Dye D2 (OO)

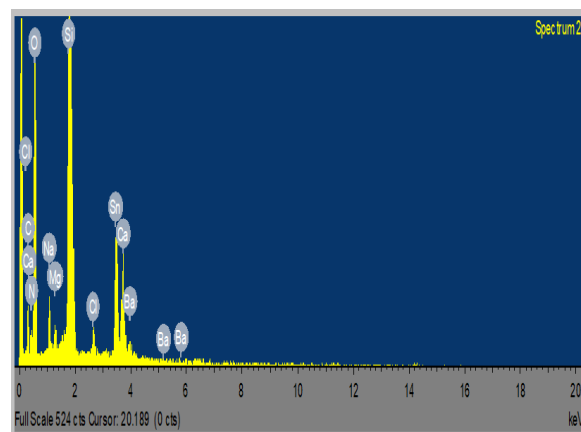


Figure 14: The EDS Spectrum of Sample 34 of BaSnO₃ Nanoparticles Sensitized with Dye 1 (LL)

3.5 FTIR Studies

Figures 15, 16 and 17 depict the schematic mid infra-red (mid-IR) spectrum of as-deposited, AD and sensitized BR, OO and LL samples of BaTiO₃ within the range of 500 to 4000 cm⁻¹ of the solar radiation. In order to ascertain the presence of the functional groups and chemical structures together with bending and stretching properties, we studied the transmittance of energy against wave number of the different dyes using FTIR. This is a characterization method that reports a set of absorption frequency/transmittance in addition to peaks displayed by its wave number which portrays functional groups and kinds of bonds existing in a biological compound (Jacox, 2003). Broad absorption band exists in the three dyes relating the presence of hydrogen bonding. All belong to the functional group of hydroxyl group. D1 has stretching vibrations; D2 has bending and stretching while D3 has stretching. D1 and D3 have several absorption bands in the entire IR spectrum indicating both are complex structures. D2 has few absorption bands hence simple molecule/structure. Finger print region is observed in all the dyes. Table 2 shows the strong absorption spectrum within 3550—3200, 2854—2926, 2855—2975, 1432—1621, 1150—911 and 858—733 cm⁻¹ which indicates bending in hydroxyl compound/O—H vibrations; stretching in Methyl group; Bending in Cyclo alkane; Stretching in Aromatic ring group; Stretching in C—O—C group and Bending in C—H group apportioned according to the obtained wave number and corresponding transmittance values depicted in figures 16, 17, and 18 for different dyes.

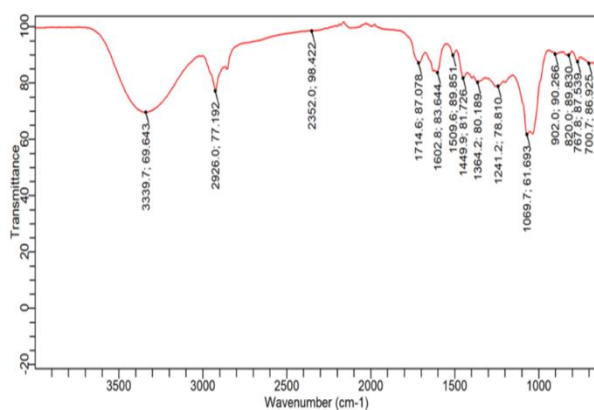


Figure 15: Mid-IR Transmittance Spectrum Regions of Henna (Laali) Dye 1

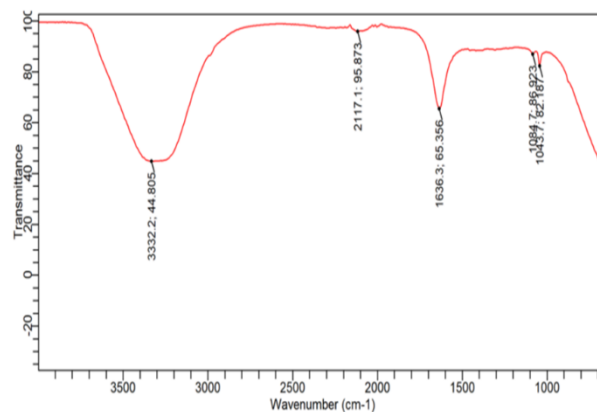


Figure 16: Mid-IR Transmittance Spectrum Regions of Dye 2 (OO)

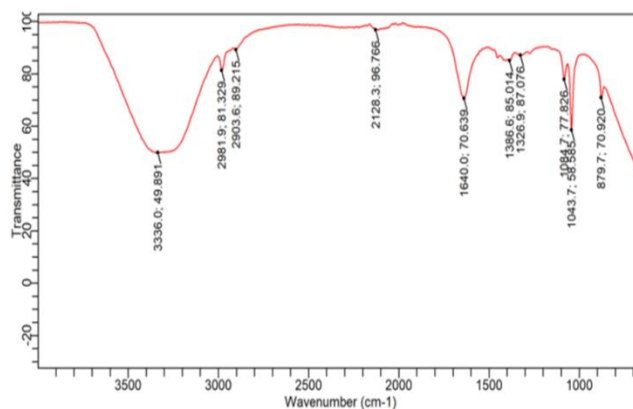


Figure 17: Mid-IR Transmittance Spectrum Regions of Beetroot (BR) Dye3

The detailed study of the FTIR showing different functional groups is depicted in table 2. The bending and stretching vibrations specific to their different and corresponding wavelengths resulted from fairly large crystallite size that led to reduced E_g rooted in dye addition. The dyes' characteristics of bending, stretching and tailing offers stability, fast/flexibility and broadening of photoluminescence peak respectively in the synthesized samples.

Table 2: FTIR investigation showing functional groups and vibrations existent

Wavenumber Range (Cm ⁻¹)	Kinds of vibrations	Functional groups
3550—3200	Bending	O—H vibrations
2854—2926	Stretching	Methyl group
2855—2975	Bending	Cyclo alkane
1432—1621	Stretching	Aromatic ring group
1150—911	Stretching	C—O—C group
858—733	Bending	C—H group
550—690	Bending	C—Cl group

Adopted from C.N. Eze IJSRPAS, Vol.11, Issue.6, pp.01-04, December 2023

4.0 CONCLUSION AND FUTURE SCOPE

The synthesis, via CBD technique, of as-deposited plus sensitized ternary nanostructures of BaSnO₃ on substrate slides followed by annealing reveals polycrystallization of the films without much significant effect on the dye-sensitized structural pattern. The thin films of the BaSnO₃ maintains a preferred orientation in the (110) plane as established by the XRD pattern. The surface studies revealed that the dyes had essential influence occurring at the surface configuration of the synthesized products outlining porous structures which is advantageous for dye loading utilizable in DSSCs. It created a large surface area for sunlight harvesting. The analysis of the optical studies involving E_g inferred that the E_g of the sensitized materials were increased in for the samples sensitized with natural dyes. All the samples have low energy band gap thereby finding their applications in window coatings. This smaller value of E_g according to our research albeit, in comparison to the standard value of 3.1 (generally, 2.9-4.2) eV could be attributed to local state formation near the band edge and other factors associated with difficulties in synthesizing bariun stannates as stated earlier. The great transmittance in the near infrared (NIR) and low E_g revealed according to the products consequently brands them decent potential products for the building of roofs and walls and window coatings of poultry structures. Due to the low reflectance of the materials, it could potentially give high efficiency if used in solar panels because it will reduce the amount of solar energy that would have reflected. This proves a potential actual significant contemplation for the building engineering. The lowered extinction coefficient of the sensitized

samples shows that the materials potentially can be used in window layer in solar cell applications.

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