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The Effect of Dopants on the Solid State and Optical Parameters of TiO_2 / MgO Thin Film

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ABSTRACT

The impact of fossil fuel on the environment, coupled with the economic crisis and environmental pollution, which is as a result of the mining of crude oil which is used in the production of fossil fuel, made the researcher to investigate into alternative areas of energy production. This work presents the successful deposition of Titanium Magnesium Oxide [TiO_2 / MgO] doped with Spinacia oleracea, Brassica oleracea and Hibiscus sabdariffa leaf extract on plain glass substrate slides using spray pyrolysis deposition techniques, solvent extraction method was used to extract the dye. This sprayed film properties were characterized for optical properties, the result of the analysis revealed an increase in absorbance within the UV region [300–350 nm], indicating strong photon absorption at shorter wavelengths. A gradual decrease in absorbance across the visible transmittance revealed troughs at the UV regions and crest at Visible and NIR. Crest and troughs observed in the reflectance spectra are attributed to thin film interference effects. The result obtained are attributed to the doping effect of thin-film. The band gap obtained are between 1.1 and 1.3 eV for the doped and 1.9 eV for the undoped. Thin films with band gap range between 1.1 - 1.3 eV corresponds with the ideal single- junction photovoltaic solar spectrum threshold and can be utilized in the manufacturing of commercial solar panels, and concentrator photovoltaics, while optical band gap of I.9 eV is optimized for harvesting high-energy photons. Its band gap suggests potential for photocatalytic and agrivoltaics applications.

INTRODUCTION

The overwhelming challenges which emerges from drilling, refining and transportation of oil, the environmental pollution that includes land, sea and air pollution, which claims thousands of lives annually and also the economic impact of all these challenges, made many researchers to pick interest in seeking alternative sources of energy, energy conversion and also storage. This incite the need for establishment of self-sustaining energy systems that will be abundant in nature, pollution free and environmentally friendly. Photovoltaic technology is being used to achieve this aim (Haloui, *et al.,* 2015). Among the devices for harnessing solar energy is solar cell. This is a device that converts sunlight directly into electricity using photovoltaic cells. These solar cells are in generations; first, second and third generations (Bhatti *et al.,*2002).

Ternary thin films, on the other hand, are materials composed of three elements and are designed to combine the properties of each constituent, resulting in unique structural, optical, and electronic characteristics. These films are often engineered to achieve properties unattainable with single or binary materials. Ternary thin films are particularly valuable in applications such as optoelectronics, catalysis, and energy storage because of their tunable properties (Liangri *et al.,* 2024).

Extract from natural leaves, roots, bark, insect (cochincal) and minerals as dye, is a coloring agent which can serve as an alternative to synthetic dyes. Natural dye is non-carcinogenic, eco-friendly and biodegradable (Pallab *et al.,* 2025). The income gotten from the cultivation of dye- producing plants, aids in the improvement in the living standard of farmers, that are involved in the cultivation of dye producing leaves or plants. (Iqbal & Ansari 2021). Dye sensitized solar cells

(DSSC), are one of the most promising devices that uses photoanode, light absorbing dye and an electrolyte for solar energy conversion into electricity. (Liviu *et al.,* 2025). The choice of any method of extracting dye from leaf depends on leaf type, dye properties, desired extraction efficiency, equipment availability and environmental consideration. The aim of this study is to mainly investigate the influence of these leave extracts on the optical and solid state properties of the studied ternary thin films. Report from related literature of ternary thin films showed that they have efficient solar energy conversion ratio and are also potential candidates for photo-electrochemical solar cells fabrication (Osuwa *et al.,* 2010). Direct band gaps is in the range of 1.30 to 2.30 eV for $CuSbS_2$ ternary thin films have been reported (Ezeugwu *et al.,* 2010). Ternary ($N1_3Pb_2S_2$) thin films by chemical bath deposition method with a band gap of 1.4-2.0 eV showed strong absorption in the visible region (Ho, 2016).

MATERIALS AND METHODS

Materials

The materials that were used in this work are all commercially available and of high-grade purity. The chemicals used were Acetone $(CH_3)_2$ CO, Distilled water, hydrogen Chloride, (HCl), Titanium (iv) oxide (TiO_2) Magnesium Oxide (MgO), Ammonia (NH_3) and Sodium Thiosulphate ($Na_2S_2O_3$), stop watch, microscopic glass slides, slide box, oven, analytical weighing balance, magnetic stirrer, Hand dryer, spatula. filter paper. UV Machine of 7565 UV-Visible spectrophotometer. Spray pyrolysis techniques was used in this work, while solvent extraction method was used for extracting the dye from *Spinacia oleracea*, *Brassica oleracea* and *Hibiscus sabdariffa* leaves. The 75 x 25 mm, 1.0 mm thick soda-lime substrates (glass slides) were purchased from Ogbete main market, Enugu state, Nigeria. The substrates were washed with water, rinsed with distilled water, soaked in 200 mL HCl for 60 mins, removed and rinsed with ordinary water, washed one after the other with detergent and foam sponge, removed and rinsed with distilled water to remove some impurities, soaked in acetone for 10 minutes and allowed to dry. *Spinacia oleracea*, *Brassica oleracea* and *Hibiscus sabdariffa* leaves were purchased from the international market in Ebonyi State, Nigeria, the leaves were washed with water, rinsed with distilled water, the water in the leaf was allowed to drain, the leaves weighed 48.0 g each. The leaves were sliced, placed in three different glass beakers labeled A, B, C and soaked in 130mL of methanol for 3 hrs, filter paper was used to filter the dye, the dye extracted was sealed with a cap and kept under room temperature. Methanol was used in the extraction of dye from these leaves because, it dissolves a wide range of compound, preserves the dye and can also evaporate easily.

Film deposition

In a 50 mL glass beaker, 7.00 mL of 3.0 M of TiO and 13.0 mL of 4.0 M of MgO_2, 0.07 mL of 0.40 M of sodium thiosulfate and 0.7 mL of 13.0 M ammonia, was added to 9.2 mL of distilled water, the mixture was stirred for 5 minutes, using magnetic stirrer to obtain a homogenous solution, following the stirring, 10 mL of the solution was measured out with a pump syringe and placed into a spray pyrolysis sample bottle, attached to the air brush's nozzle valve rod. The solution was sprayed for 60 secs onto heated substrate (glass slide), using an electrical hot plate to heat the substrate to 300^{o}C to facilitate decomposition. The thin film was left as control for this work, after undergoing annealing under 200 ^{o}C.

The following chemicals were synthesized in a glass beaker: 7.00 mL of 3.0 M of TiO and 13.0 mL of 4.0 M of MgO_2, 0.07 mL of 0.40 M of sodium thiosulfate and 0.7 mL of 13.0 M ammonia was added to 9.20 mL of distilled water, making the total quantity of precursor produced 20 mL, the mixture was stirred for 5 minutes using magnetic stirrer to obtain a homogenous solution, after the stirring, 10 mL of the precursor was measured using pump syringe and doped with 4 mL of *Spinacia oleracea*, *Brassica oleracea* and *Hibiscus sabdariffa* extract separately, it was vigorously stirred for 5 minutes using magnetic stirrer to obtain a homogenous solution, the precursor was placed into a spray pyrolysis sample bottle, attached to the air brush's nozzle valve rod. The solution was sprayed for 60 secs onto heated substrate (glass slide), using an electrical hot plate to raise the substrate temperature to 300 ^{o}C, the samples were annealed at 200 ^{o}C respectively. UV Machine of 7565 UV-Visible spectrophotometer was used for the characterization of the thin films to determine its optical properties and it's possible application.

RESULTS AND DISCUSSION

UV analysis

UV-VIS spectroscopy is considered as the most important spectrophotometric technique that is most widely used for the analysis of variety of compounds. This technique works on the basis of the measurement of interaction of electromagnetic radiations [EMR] with matter at particular wavelength. (Onah *et al.,* 2023). This radiation, has a spectral range approximately around 190–800 nm, which also differ in terms of energy ranges, and type of excitation from other related regions (Agbo *et al.,* 2021).

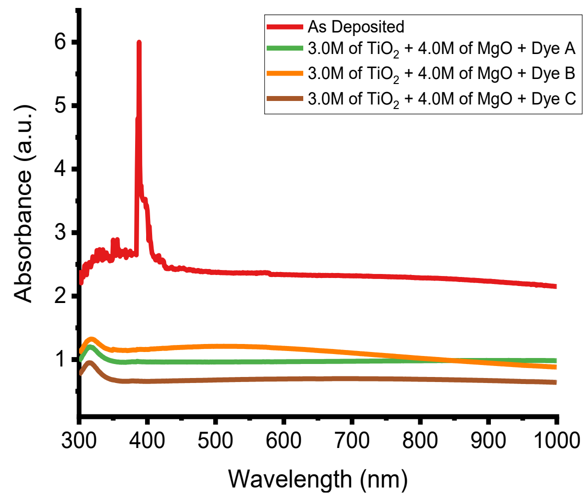

Figure 1: A graph of Absorbance against wavelength

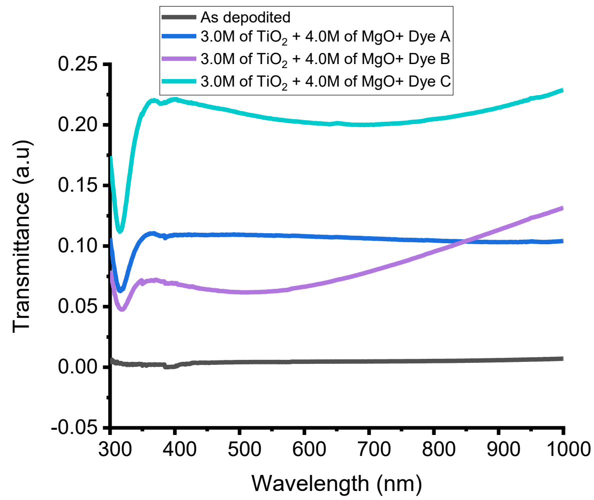

Figure 2: A graph of transmittance against wavelength

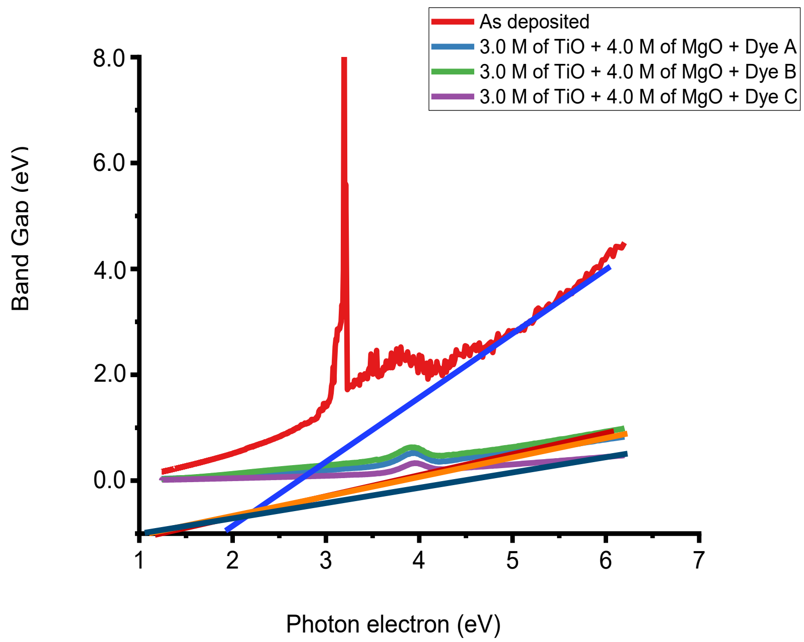

Figure 3: A graph of band Gap against photon energy

Analysis of the optical properties of TiO_2 / MgO

B16 represents 3.0 M of TiO_2 + 4.0M of MgO as deposited

B17 represents 3.0 M of TiO_2 + 4.0M of MgO + *Hibiscus sabdariffa*

B18 represents 3.0 M of TiO_2 + 4.0M of MgO + *Brassica oleracea.*

B19 represents 3.0 M of TiO_2 + 4.0M of MgO + *Spinacia oleracea.*

Analysis of the Absorbance of B16

The absorbance spectrum of sample B16 as shown in Figure 1, exhibited a gradual increase within the wavelength range of 300 – 400 nm, followed by a sharp reduction around 450 nm. Beyond this region, the absorbance remained nearly constant from 450 nm up to 1000 nm.

The enhanced absorbance response in the UV region indicates strong interaction with high-energy photons, while the sudden decline suggests the presence of electronic transition thresholds within the film structure. The relatively stable absorbance in the VIS – NIR region implies consistent optical transparency over a broad spectral range, such characteristics make this thin film suitable for optical coatings requiring controlled UV – VIS transition behaviour, particularly in applications such as glare-reducing surfaces, sensors, laser components, and camera systems. These findings agree with earlier reports by (Onah *et al.,* 2023 & Agbo *et al.* 2021).

Absorbance Behaviour of Dye-Doped Films (B17 – B19)

The dye-doped thin films (B17, B18, and B19) demonstrated noticeable absorbance enhancement around 320 nm as also shown in Figure 1. Absorbance values of approximately 0.8 a.u., 1.0 a.u., and 1.4 a.u. were recorded for *Spinacia oleracea*, *Brassica oleracea* and *Hibiscus sabdariffa* doped samples, respectively, this indicates that dye incorporation modifies photon absorption in the UV region through additional electronic states introduced into the oxide matrix. Furthermore, a stable absorbance response was maintained between 350 nm and 650 nm, confirming uniform transparency within the visible spectrum.

Between 800 nm and 950 nm, samples B17 and B18 exhibited closely related spectral behaviour, suggesting similarities in their optical transition processes. These results highlight the potential of the films for VIS – NIR transparent coatings and wavelength-selective optical filters. Comparable trends have been reported by (Igwuoko *et al.* 2025 & Derdakh *et al.* 2019).

Transmittance analysis of Sample B16

The transmittance response of sample B16 – 19 as displayed in Figure 2 is discussed thus:

the transmittance spectrum of sample B16 remained almost constant at approximately 0.1 across the entire wavelength range of 300 – 1000 nm. This behaviour indicates that the film exhibits very low optical transmission, suggesting significant absorption or scattering effects within the oxide structure. Such reduced transmittance implies that the film may be suitable for applications requiring strong light attenuation or optical shielding. Similar observations have been documented by (Naveensubramaniam *et al.,* 2023 & Manikandan *et al.,* 2017).

Transmittance Response of Sample B17

Sample B17 showed an initial reduction in transmittance around 310 nm, followed by an increase near 360 nm. Thereafter, the transmittance remained stable from 400 nm to 1000nm. The improvement in transmission around 360 nm may be attributed to optical modifications induced by Zobo extract doping. Additionally, the suppressed UV transmission near 310 nm indicates potential UV – blocking capability, making the film useful for UV – protective coatings, polymer stabilisation, and skin protection applications, these findings agree with earlier reports by (Melford *et al.,* 2019 & Panuwat, 2020).

Transmittance Response of Samples B18 and B19

Samples B18 and B19 displayed more complex transmission behaviour. After an initial reduction near 310 nm, transmittance increased up to 360 nm, followed by another decrease around 550 nm. A gradual increase was then observed toward longer wavelengths.

This spectral variation reflects the influence of *Spinacia oleracea* and *Brassica oleracea* which introduce additional absorption bands and modify defect-related electronic states. Such optical tunability is advantageous for tailoring spectral windows in optical filtering and detection systems. These findings align with (Murcia *et al.* 2020 & Featherstone *et al*., 2015).

Band gap Analysis

Figure 3 shows the optical band gap energies of the TiO_2 / MgO (3.0 M TiO_2 + 4.0 M MgO). The undoped and dye-doped samples exhibited different band gap values, the band gaps of doped samples are within 1.1 - 1.3eV while the band gap for the undoped sample is 1.9eV. Thin films with the band gap range between 1.1 - 1.3 eV corresponds with the ideal single- junction photovoltaic solar spectrum threshold. These band gap range, maximizes sunlight absorption and minimizes thermal energy losses and can be utilized in the manufacturing of commercial solar panels, bottom tandem layers, concentrator photovoltaics and flexible solar sheets, this finding is in agreement with the findings of (Song, *et al.,* 2019 & Bush, *et al.,* 2017).

Material with optical band gap of I.9 eV which is for undoped TiO_2 / MgO is optimized for harvesting high-energy photons, which serves as a top cell in multi-junction tandem devices.This materials are typically used in photovoltaic cells, LEDs, or other devices where absorption or emission of light is crucial. Its band gap suggests potential for photocatalytic applications, such as water splitting or pollutant degradation, it can also be used in Agrivoltaics. The result can be compared with the results obtained by (Oleksiy *et al.,* 2022, Ballestín-Fuertes, *et al*., 2021 & Zhang, 2020).

CONCLUSION

This study demonstrates successful synthesis and optical characterisation of TiO_2 / MgO thin films using spray pyrolysis. The incorporation of natural dye extracts from *Spinacia oleracea*, *Brassica oleracea* and *Hibiscus sabdariffa* leaves effectively modified the optical properties of the thin films. The band gaps of the doped samples are within 1.1 - 1.3eV while the band gap for the undoped sample is 1.9eV. Thin films with this doped band gap range, maximizes sunlight absorption and minimizes thermal energy losses and can be utilized in the manufacturing of commercial solar panels, bottom tandem layers, concentrator photovoltaics, while materials with 1.9eV band gap can be optimized for harvesting high-energy photons, which serves as a top cell in multi-junction tandem devices and can be utilized in photovoltaic cells and agrivoltaics among other applications. The use of natural dyes as dopants offers an environmentally friendly and cost-effective approach to tailoring thin-film optical properties.

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