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Comparative characterizations of plant and mineral dye as potential photosensitizer in Dye-Sensitized Solar Cell

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ABSTRACT

The advent of dye-sensitized solar cells (DSSC) has expansively seen more studies as an alternative to silicon-based and thin-film solar cells due to their simple structure and relatively low production cost. In dye-sensitized solar cells, the dye is one of the major components for high power conversion efficiencies. The extracted dyes were characterized by powder x-ray diffraction, x-ray fluorescence, Scanning electron microscopy, UV-visible spectroscopy, and Gas Chromatography-Mass spectrometry analysis. The structure of the mineral dye contains constituents that enhance better absorption of solar radiation for use in a Dye-Sensitized Solar Cell (DSSC). The calcium and iron content of the mineral dye studied as revealed by the mineralogical analysis done using the powder x-ray diffraction (XRD) and the x-ray fluorescence (XRF) suggest this. These metals serve as bounding complexes to other organic components in the dye, like in the case of Ruthenium complexes dye for efficient absorption of solar radiation, especially in the visible light region. The functional groups present in the dye also confirm what favors good absorption of solar radiation for DSSC application. The Organic chemical compositions present in the mineral dye which were obtained by the Gas Chromatography-Mass spectrometry analysis also confirm the functional groups of carbonyl, Amine, and hydroxyl revealed by FTIR, which were responsible for solar radiation absorption. There are strong absorption narrow bands in the visible region with peaks at around 424 nm (2.903 a.u) and 486 nm (2.973 a.u). Optical band gaps of 1.66 eV and 2.27 eV for the mineral dye and plant dye respectively, were obtained. The properties of the dyes studied give potential substitutes to the relatively expensive ruthenium-based dyes for use in DSSC fabrication.

Keywords: Photosensitisers, Mineral, Dyes, Photovoltaic, Plant dye.

1. INTRODUCTION

The birth of dye-sensitized solar cells (DSSCs) in the year 1991 has extensively witnessed more studies as an alternative to both crystalline and amorphous silicon-based solar cells, and the thin film solar cells. This is because of their simple structure, flexibility, and relatively low production cost (Akinsola et al., 2021a & b; Monzir et al., 2015). With the advantages this device portrays, its low efficiency compared to that of silicon-based cells has limited its commercial implementation (Jun et al., 2014). The third-generation photovoltaic device, known as DSSC, can convert photons

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to electrical energy at power conversion efficiency of about 13% (William et al., 2017). A huge effort is continuously going into the optimization of the various components of DSSC with the sole aim of fabricating improved efficient and stable cells.

Dye-sensitized solar cell is made of a fluorine-doped Tin Oxide front contact (FTO) on glass, nanoparticles semiconductor photoanode covered with sensitizing dye, a hole conducting electrolyte, and platinum or graphite coated FTO back contact (Akinsola et al., 2023). The dye is one of the principal components for high conversion efficiencies of a DSSC. Lately, noticeable developments have been made in the engineering of dye structures to enhance the performance of the device.

It was also affirmed by Ghann et al. (2017), that the sensitizers currently used in the fabrication of solar cells are transition metal coordination complexes such as Ruthenium (II) carboxylated polypyridyl complexes, because of their intense charge-transfer absorption in the whole visible range and highly efficient metal-ligand charge transfer transition (MLCT). However, Natural dyes are more desirable than these synthetic dyes because they are more economical, easily attainable, and abundant in supply and environmentally friendly. In addition, they are discovered to have large absorption coefficient owing to allowed π to π^* transitions. These pigments are derived from various plant parts such as flower petals, leaves, roots and fruits pulp/bark. The relatively quick degradation of even the natural dyes obtained from plants as compared with the metal coordination complexes calls for considering of an alternative natural dye with cost effectiveness and good stability. Natural dye can be categorized into plant and mineral dyes. In this paper, comparative study of the properties of dyes obtained from natural mineral (which is not yet being widely used for DSSC) and a root vegetable spice was carried out and their suitability in absorbing solar radiation for excitation of electrons in generating electricity via Dye-sensitized solar cell is considered. This is expected to be a novel panacea to the quick degradation challenge of the use of natural plant dyes in fabricating DSSC.

2. METHODOLOGY

2.1. Materials

The rock-like mineral, yombotumtum (YT), was obtained from the southern part of Ghana, Western Africa. The yombotumtum is being used by majority to dye hair in some parts of Africa.

Its origin was traced to a region in Ghana, and it was learnt that the name is of the Ghanaian Language (Adebayo et al., 2006). The plant dye is from *Curcuma longa* (Turmeric).

2.2. Methods

The dyes were separately extracted from the powder using ethanol, with decantation technique. The extraction technique used is by crushing of the dye materials into powdery form and soaked in an organic solvent; ethanol precisely, to efficiently extract the dye content from the soaked materials. The structural properties of the dyes were studied by carrying out X-Ray Diffraction (XRD) using the X-Ray Diffractometer (Rigaku D/Max-IIIC, PW 1800) and Scanning Electron Microscopy (SEM). The quantitative analyses of the dyes were done using the X-Ray Fluorescence (XRF) technique, to obtain the elemental composition of the dyes. The functional groups present in the dyes were determined using the Fourier Transform Infrared (FTIR) Spectroscopy. The Absorption spectra of the dye was studied within the visible region of the electromagnetic radiation, and it was done using the UV-visible Spectrophotometer (VWR: UV-6300PC Double Beam Spectrophotometer).

The dye's organic compositions were studied using the Gas Chromatography-Mass Spectrometer (Agilent Technologies – 5975C Inert MSD with Triple Axis Detector). Helium was used as carrier gas. Comparative analytical method was employed by comparing the given spectrum to a spectrum library to see if its characteristics are present for some sample in the library. MSD – Mass selective detector was used. NIST – National Institute of Standards and Technology's Library Database was considered and compared.

3. RESULTS AND DISCUSSION

3.1. Optical Properties

Absorption of electromagnetic radiation is the process by which certain energy is being taken up with photon by matter. The absorption spectra of yombotumtum dye are given in figure 1. Electromagnetic spectrum comprises of Radio wave, Infrared, Visible light, Ultraviolet, X-Ray and Gamma Ray. The interest of this work is in visible region since the dye is being studied as a potential photosensitizer in a Dye-Sensitized Solar Cell (DSSC) which absorbs solar radiation within the visible region of the electromagnetic radiation.

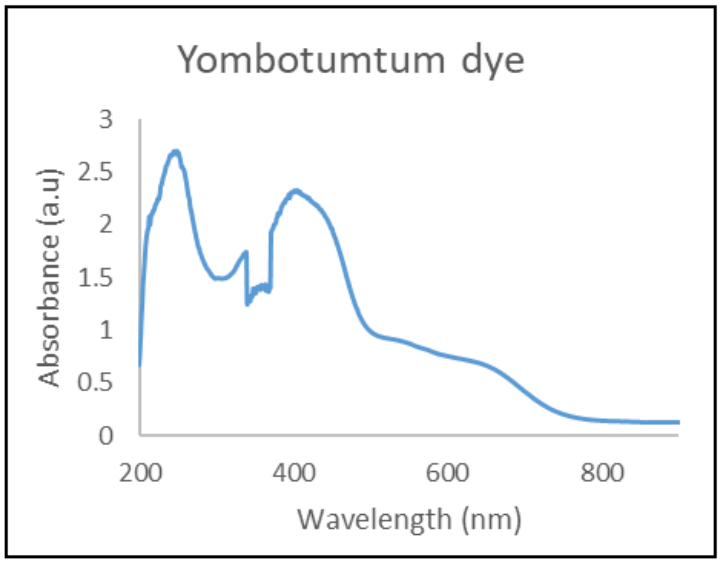

Figure 1. Absorption Spectra of yombotumtum Dye.

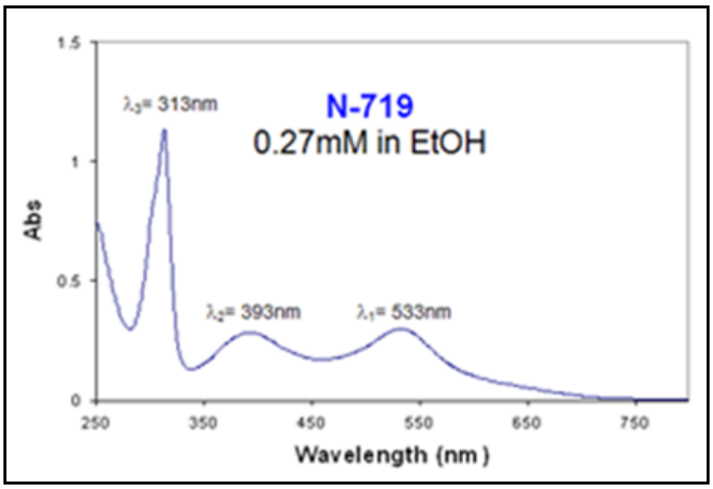

Figure 2. Absorption Spectra of Ruthenium-based dye, N-719 (Product No. 703214) (Hans and Yanek, 2017).

Figure 1 indicates that the dye has a high absorption of solar radiation within the visible region. Considering figure 2, the absorption of solar radiation of a typical Ruthenium-based dye (a synthetic dye) is relatively low with respect to this mineral dye; yombotumtum (a natural dye) which shows a higher absorption (Fig 2). It is indeed a potential photosensitiser in a DSSC. The dye extract exhibited a strong absorption broad band in the visible region with peaks at around 407 nm (2.299 a.u) and 537 nm (0.905 a.u). This intense absorption in the visible region especially at 537 nm matches with what has been reported for anthocyanin and is the reason for the efficient

harvesting of photons in Natural DSSC. Anthocyanin is a group of naturally occurring phenolic compounds responsible for the colour of many flowers and fruit.

Ruthenium-based dye exhibit ligand-centered charge transfer (LCCT) transitions (π - π*) as well as metal-to-ligand charge transfer (MLCT) transitions (4d - π*) that can be observed in the absorption spectra of N-719 dye (Figure 2). The absorption bands at lower energies represent the MLCT transitions (λ_1 and λ_2) whereas the more energetically demanding transitions (λ_3 and λ_4) correspond to LCCT transitions. Promotion of an electron from π –bonding orbital to an antibonding π orbital* is denoted by π - π* transition. Section of molecules which can undergo such detectable electron transitions can be referred to as chromophores since such transitions absorb electromagnetic radiation (light), which may hypothetically be perceived as colour somewhere in the electromagnetic spectrum.

The absorption spectra of Yombotumtum dye given in figure 1 shows absorption bands at more energetically demanding transitions which correspond to LCCT transitions within the visible region. Hence, favoring a good absorption of solar radiation for the operation of a solar cell.

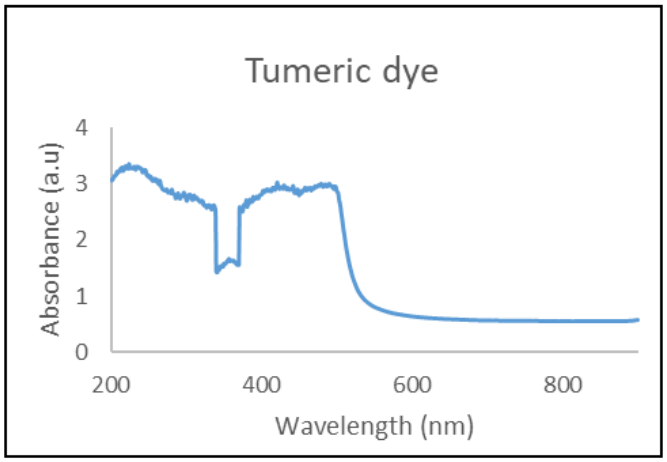

Figure 3. Absorption spectra of turmeric dye.

Figure 3 shows the absorption spectra of the plant dye studied. There are strong absorption narrow bands in the visible region with peaks at around 424 nm (2.903 a.u) and 486 nm (2.973 a.u). It is obviously of higher absorption in the visible region than the mineral dye (considering the comparison in Fig 4). The absorption peak in the ultraviolet region (about 200 nm – 370 nm) of

the electromagnetic radiation in this dye could be due to the presence of flavonoids in the plant naturally. However, it has a demerit of quick degradation relative to the mineral dye.

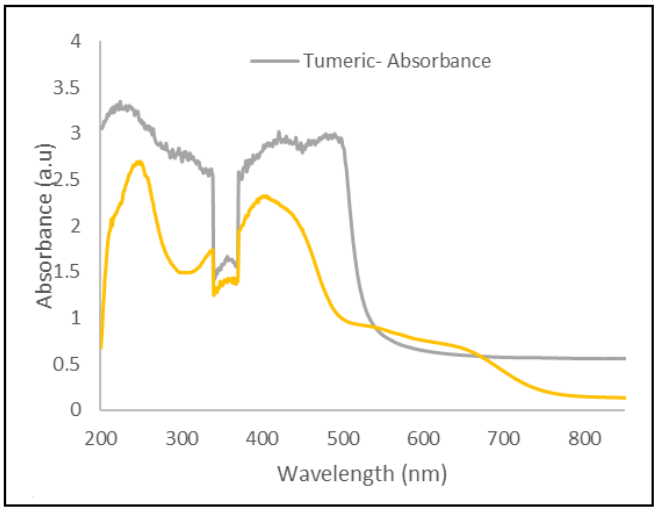

Figure 4. Comparison of the absorption spectra of turmeric and yombotumtum dye.

3.1.2. Optical Band Gap of Dyes

The optical energy band gap of the dyes was obtained from the expression,

$$E_g = (hc/\lambda_{th}) \quad \text{(Ossai et al., 2020)} \text{ ----------(1)}$$

Where, $\mathbf{E_g}$: the Band gap, **h** : Planck's constant, $\boldsymbol{\lambda_{th}}$: threshold wavelength, **c** : speed of light.

Table 1. Optical band gap of dyes with their respective offset absorption wavelengths.

Dye	***Offset absorption wavelength (nm)***	***Band gap, E_g (eV)***
Turmeric	548	2.27
Yombotumtum	750	1.66

Table 1 shows the estimated band gap for the two dyes. From the offset absorption wavelength, which is also the threshold wavelength, for the mineral dye, it is observed that it has a wider absorption range in the visible region than the plant dye. This result agrees with the report of Ade et al. (2020), which state that one way to improve DSSC efficiency is by expanding the range of light absorption from dye; become near the Near Infrared (NIR) area, which is around 940 nm. The estimated band gap for the mineral dye, as seen in table 1 shows that, even though Turmeric has higher absorption than the mineral dye, it can't perform better than the

yombotumtum, because of the later smaller optical band gap. Electrons will jump quicker from the Highly Occupied Molecular Orbital (HOMO) to the Lowest Unoccupied Molecular Orbital (LUMO) in yombotumtum dye than the Turmeric dye. The result for yombotumtum is 1.66 eV which was supposed to be due to the use of ethanol for its extraction.

3.1.3. Absorption Coefficient of Dyes

The absorption coefficient of the dyes was obtained by the use the equation 2.

$$\alpha = {}^{4\pi k}/_{\lambda} \qquad \text{----------(2)}$$

Where, α = absorption coeff., λ = peak absorbance wavelength, k = Boltzmann's Constant (in eV).

The absorption coefficient characterizes how far into a material; the light of a particular wavelength can penetrate before it is absorbed. Table 2 shows the estimated absorption coefficients of the two dyes. The mineral dye has a higher absorption coefficient that the plant dye.

Table 2. Absorption coefficient of dyes.

Dye	***Wavelength of peak Absorbance (nm)***	***Absorption coefficient (k m^{-1})***
Turmeric	495	2.189
yombotumtum	413	2.624

3.1.4. Functional Groups of the Dye

An FTIR spectrum of the yombotumtum dye is shown in figure 5. The functional groups present in an organic dye responsible for the absorption of solar radiation are the Amine, hydroxyl and the carbonyl groups. In addition to the high absorption coefficient in the visible region of the electromagnetic spectrum, the presence of hydroxyl (-OH) and carbonyl (C=O) anchoring groups in the mineral dye as revealed by the stretching vibrations at 3337.8 cm^{-1} and 1653.1 cm^{-1} respectively will enable their adsorption unto the surface of semiconductor to be used in a DSSC. The presence of the Amine group in the dye is revealed by the vibration at 3337.8 cm^{-1}. For the Turmeric, we have 1653.1 cm^{-1} and 3365.8 cm^{-1} representing the vibrations for the carbonyl and amine group respectively (see Fig 6). Krisnamurti et al. (2015) reported similar functional groups in certain related organic compounds. The absorption bands for bending vibrations are typically found in the fingerprint region (1400 – 600 cm^{-1}). These vibrations correspond to the likely metal-bonded compounds present in the region which the mineralogical analysis carried out using the X-

ray Diffraction (XRD) technique revealed. Stretching vibrations of C–O–C esters demonstrate peaks at 1086.5 cm^{-1}. These functional groups confirm the presence of chlorophyll and anthocyanin in plants dyes. Flavonoids in the plant dye is in the wavelength range of 310-350 nm which is of Phenolic compounds and confirms by the hydroxyl group. Presence of amine and carbonyl groups, especially, in a dye favor absorption of solar radiation; it favors the pi-pi* molecular electronic transition.

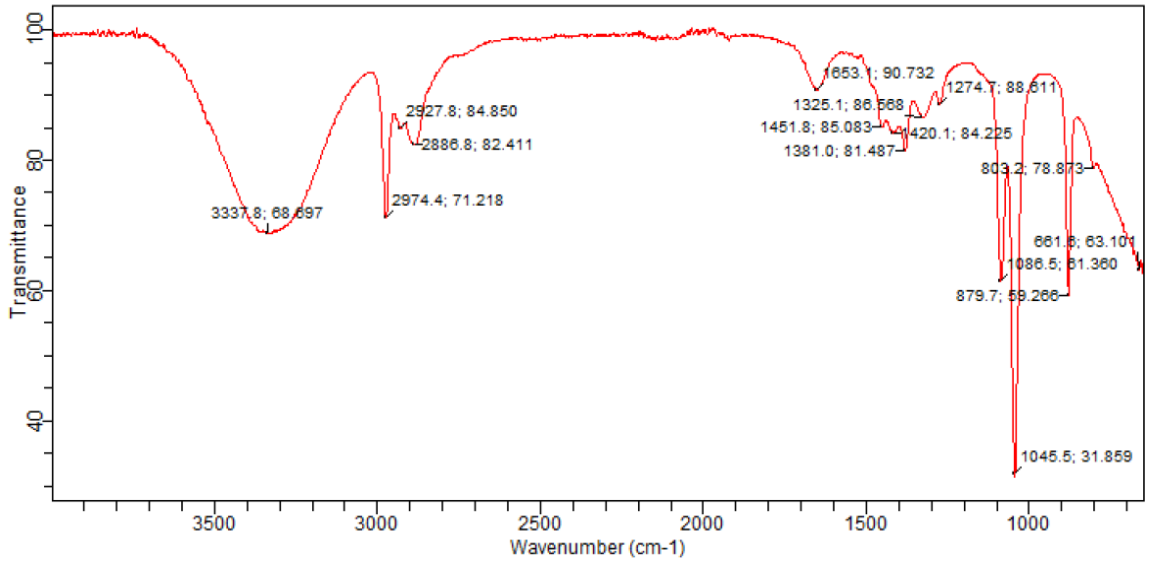

Figure 5. FTIR spectra of yombotumtum dye.

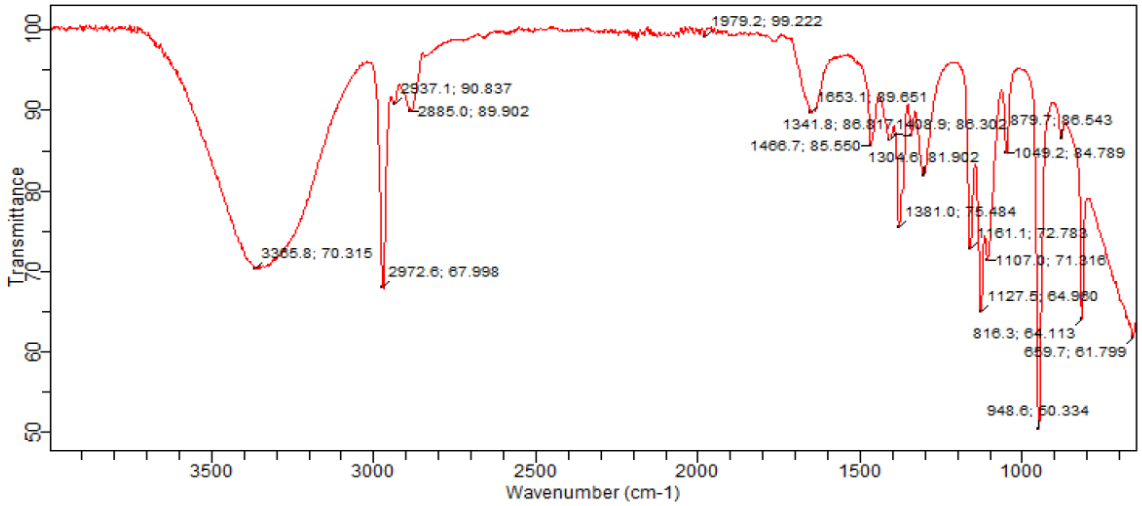

Figure 6. FTIR spectra of turmeric dye.

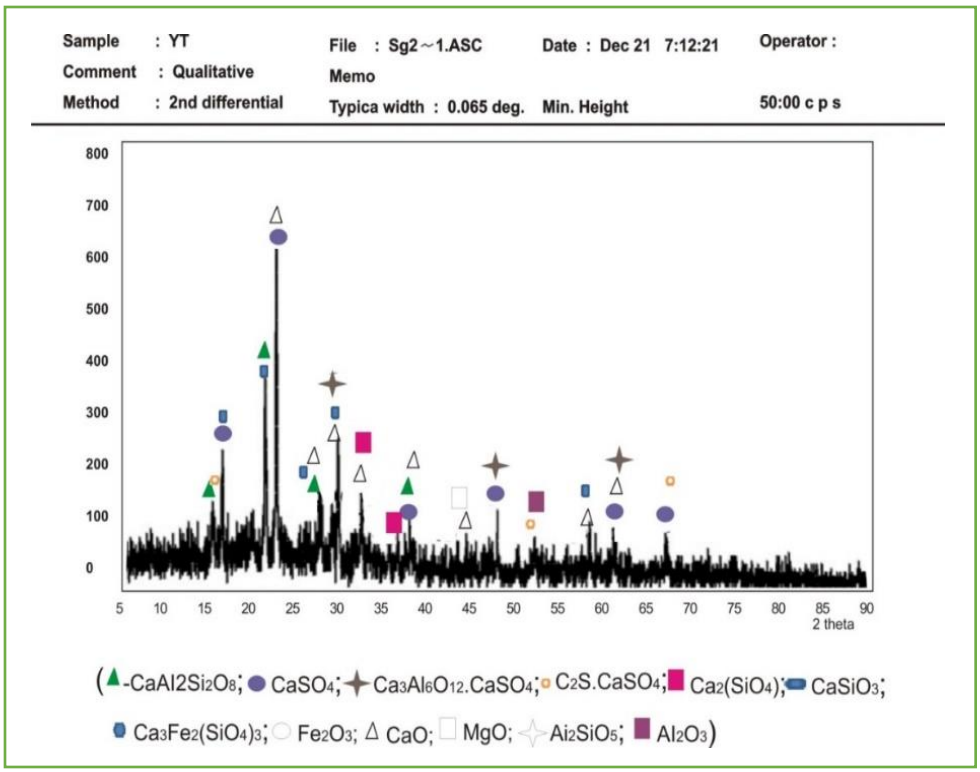

Figure 7. XRD Pattern for yombotumtum (YT) (LAB.1).

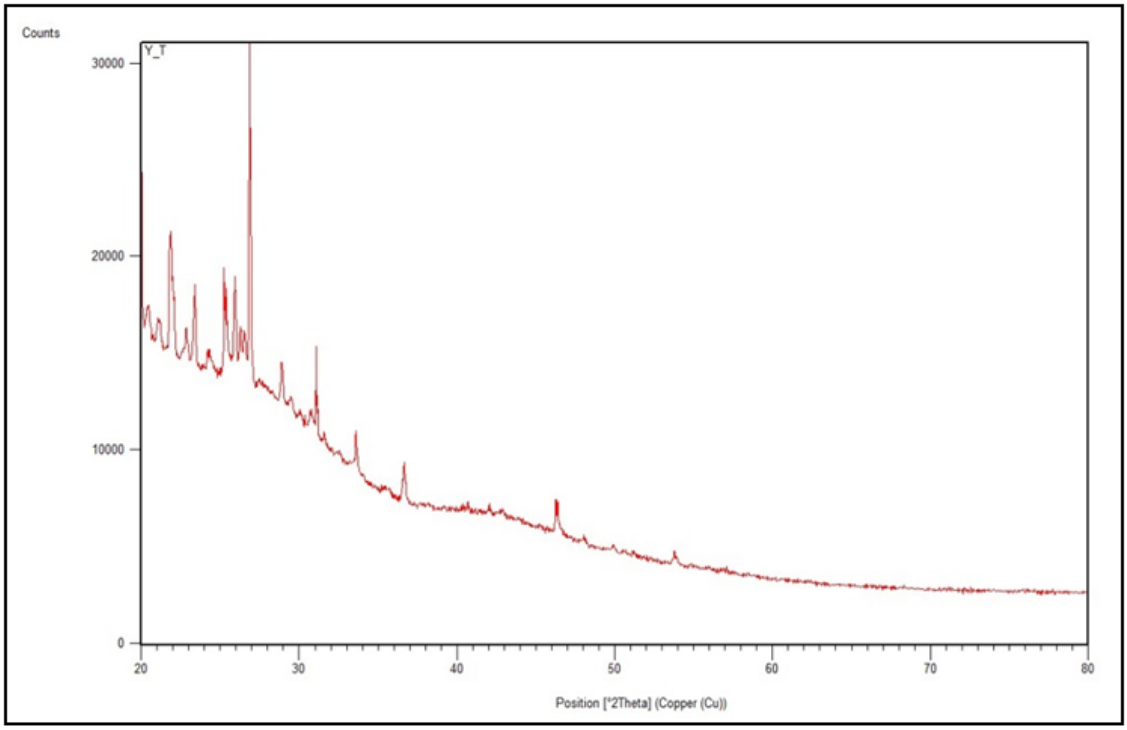

Figure 8. XRD Pattern for yombotumtum (YT) (LAB. 2).

3.1.5. Structural characterization and morphology of the mineral dye

The dye was subjected to X-ray Diffraction to obtain the mineralogical content of the materials. The idea of obtaining the mineral content is to suggest to us the compounds that make up the dye and hence the significance of them in contributing to absorption of solar radiation in a photovoltaic device. As Ruthenium metal is bounded with some groups to form a complex being used as a dye in a DSSC, so also the XRD pattern of the dye shows the presence of metal-based compounds as minerals in the dyes.

Figures 7 and 8 give the patterns of diffraction of X-ray for yombotumtum dye. It was observed that the peak with the highest intensity corresponds to $CaSO_4$ and CaO. $CaSO_4$ is the commonest form of natural sulphates and Gypsum. The next to this is the $CaAl_2Si_2O_8$ (see Fig 7) which is the chemical formula of a mineral - Anorthite, found in mafic igneous rocks.

There is no exact diffraction data file for the dye material under study, being a natural mineral that has not been worked on by scientists, to the best of our knowledge as at the time of writing this article. No JCPDS card number of a material can be used to interpret the X-Ray diffraction pattern of the dye obtained precisely. Although, the JCPDS Card no [41 - 0224] for a hydrated $CaSO_4$ was used to observe matching with the obtained XRD pattern and it was seen that the 2θ values of about 23.5^0, 28.9^0, 29.11^0 correspond to the $CaSO_4$, $CaAl_2SiO_2O_8$ with mixture of $Ca_3Fe_2(SiO_4)_3$ and Fe_2O_3, hence the need to compare figures 7 and 8 (XRD done at two different laboratories). The composition suggests a combination of different minerals, though with Calcium (Ca) compounds being the dominant and Iron compound to be considered also.

The mineral dye shows crystallinity with a few distinct peaks in the XRD pattern. The grain size of the mineral dye powder was obtained using equation (3) of Debye-Scherrer:

$$g = k\lambda / (\beta \cos \Theta) \quad \text{---------- (3)}$$

Where, λ = 1.541874 Å (wavelength of the X-ray used), β = the broadening of diffraction line measured at the half of its maximum intensity.

From figure 7, the estimated average grain size, g = 7.2 nm. The Anorthite and sulphates traces in the dye are responsible for its grayish and brownish colour. This can be justified by the connection the mineralogical content has with gypsum and being earthy. Average grain size for $CaSO_4$ (as estimated from Debye Scherrer equation) is approximately 33.2 nm.

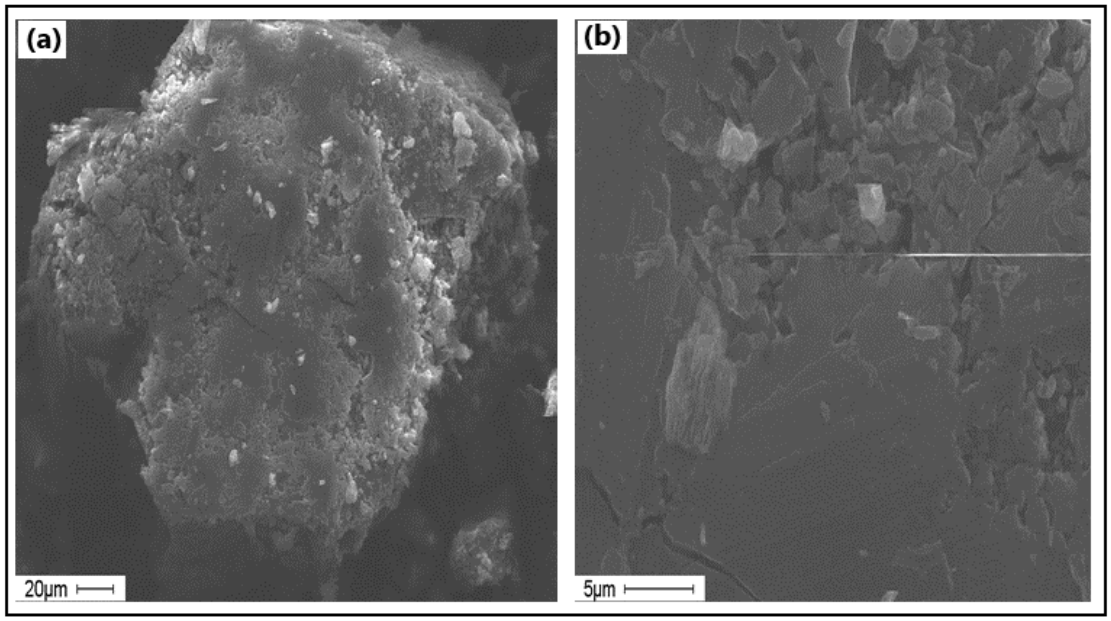

Figure 9. SEM Images of yombotumtum dye powder at different magnifications.

The Scanning electron micrograph is represented by figure 9. The two SEM images were at different magnifications. Both micrograph of the dye show a clustering of the particles of the dye. The surface morphology of the dye reveals a uniform structure and agglomeration of the constituents, together with presence of surface cracks, of the dye material. The clustered morphological observation of the dye could suggest the suitability of the dye in being better adsorb on the surface of the semiconductors to be used in a dye-sensitized solar cell. Since one of the desired morphological properties of a TiO_2 or ZnO to be used in a DSSC is the good porosity which will favour better dye adsorption, hence a uniformly agglomerated particles in a dye material will provide an excellent adsorption unto the semiconductor in the organic solar device.

3.2. Quantitative Analysis

The elemental composition of the yombotumtum dye was summarized in Table 3. From the analysis it was observed that the elements with the prominent concentrations in the dye material are Potassium (K), Calcium (Ca) and Iron (Fe) with 1552.830, 366.4 and 1009.9 ppm respectively. The high concentration of Ca and Fe in the dye could be because it has its source from the earth (being a natural mineral). Also, the iron in the dye material is responsible for its brownish green colour, owing to the magnetite features which is the natural ore of iron. This, consequently,

eventually enhances its high absorption of electromagnetic radiation in the visible region. The results discussed under the optical properties and as seen in table 4 justify this fact and as revealed by the mineralogical content given by the XRD result.

Table 3. Elemental composition of yombotumtum.

Element	***Concentration (ppm)***	***Element***	***Concentration (ppm)***
K	1552.830 ± 492.552	Ni	200.757 ± 23.242
Ca	366.405 ±56.886	Cu	292.509 ± 30.924
Sc	< 71.832	Zn	211.576 ± 13.234
Ti	90.469 ± 28.218	Ga	< 20.335
V	86.275±26.952	Pb	< 21.818
Cr	94.831±18.211	Se	< 25.691
Mn	< 44.811	Br	42.734 ± 8.073
Fe	1009.944 ± 19.513	Rb	< 42.443
		Sr	< 48.171

3.3. Gas Chromatography-Mass Spectroscopic Analysis of the mineral dye

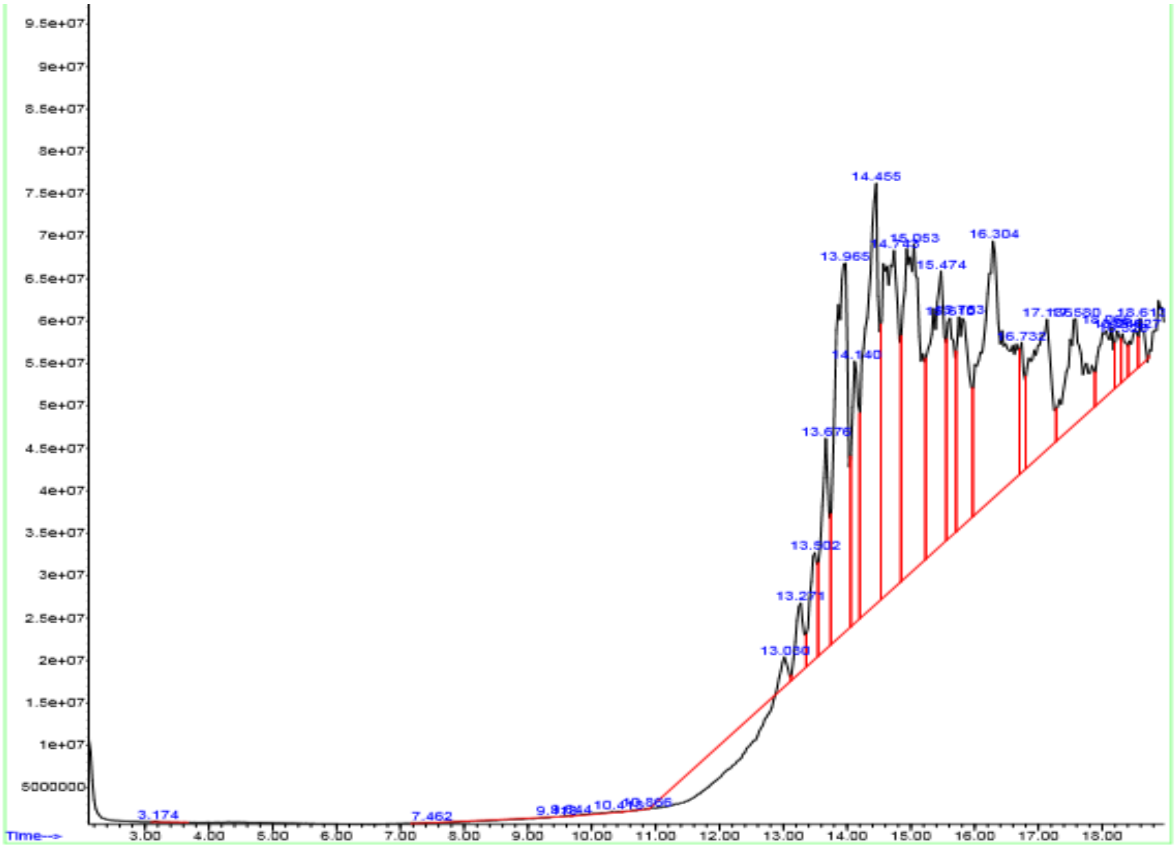

Figure 10. Yombotumtum chromatogram.

The chromatogram of the mineral dye under study is presented in figure 10. By comparing the obtained spectrum with the NIST library database, the analysis of the organic compounds present in the material are summarized in table 4. The essence of this is to ascertain the chemical compositions which are possibly responsible for absorbing solar radiation for excitation of electrons in the dye for application in DSSC.

Table 4. Analysis of Organic Compound Composition in yombotumtum.

Dye sample	***Abundance on the Chromatogram***	***Retention Time (RT) on Given spectrum (Mins)***	***Retention Time (RT) on spectrum Library (Mins)***	***Substances/Compound present***
Yombotumtum	7.7E+7	14.455	14.454	i. Dodecanoic acid, 2,3,4,6 – tetrachlorophenyl ester ii. Dodecanoic acid, 1,2,3 – propanetriyl ester iii. 4 – Nitrophenyl laurate
	7.0E+7	16.304	16.314	i. 4-Dibenzofuranamine ii. Dodecanoic acid, 1,2,3 – propanetriyl ester iii. 2H – pyrimido [2, 1-b] [1,3] thiazin – 6-one, 8-amino – 3, 4 – dihydro-

The ester family (C = O stretch) in the mineral dyes is expressing the carbonyl stretching absorption. That is, Dodecanoic acid, 1,2,3 – propanetriyl ester ($C_{39}H_{74}0_6$) in the mineral dyes aid good absorption of solar radiation in the visible region. Dodecanoic acid, 2,3,4,6 – tetrachlorophenyl ester confirms the hydroxyl group observed via the FT-IR spectroscopy. 4-Dibenzofuranamine ($C_{12}H_9NO$) is most likely for the amine group seen in the mineral dyes. All these are suspected to be responsible for the behaviour of the dye with light. Hence, a potential photosensitizer than can even be optimized for enhancement of DSSC efficiency.

4. CONCLUSION

In this work, the properties of a mineral dye in comparison with a plant dye is reported. The characteristics studied revealed how suitable the dyes are in being efficient photosensitizer in a Dye-sensitized solar cell (DSSC). There are strong absorption narrow bands in the visible region

with peaks at around 424 nm (2.903 a.u) and 486 nm (2.973 a.u). The optical band gaps of the mineral and plant dyes are 1.66 eV and 2.27 eV respectively. These energy gaps are such that will absorb visible light of solar radiation. The presence of hydroxyl (-OH) and carbonyl (C = O) anchoring groups in the mineral dye as revealed by the stretching vibrations at 3337.8 cm^{-1} and 1653.1 cm^{-1} respectively will enable their adsorption unto the surface of semiconductor to be used in a DSSC. The presence of the Amine group in the dye is revealed by the vibration at 3337.8 cm^{-1}. For the turmeric, we have 1653.1 cm^{-1} and 3365.8 cm^{-1} representing the vibrations for the carbonyl and amine group respectively. Although the studied mineral and plant dye are being used for different purposes for decades, it is discovered to possess tendencies of being a good absorber of solar radiation in the visible region of electromagnetic radiation, and a promising substitute to the relatively expensive and easily-degrading dyes commonly in use for DSSC at present.

5. ACKNOWLEDGMENTS

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6. CONFLICT OF INTEREST

There is no conflict of interests.

7. REFERENCE

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