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Fabrication and Comparison Performance of Dye-Sensitized Solar Cells Based on Single and Cocktail Natural Dyes

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ABSTRACT

Three natural dyes were used in the fabrication of Dye Sensitized Solar Cells (DSSCs): anthocyanin dye from pride of barbados flower and chlorophyll dyes from masquerade tree leaf and pumpkin leaf dye extracts. The chlorophyll and anthocyanin dyes were hybridized in a cocktail in ratio 1:1 by volume. The purpose of the research is to fabricate enhanced DSSCs based on natural dye at low cost.The photoanode was fabricated using P25 TiO_2 powder coated on the fluorine doped tin oxide (FTO) conducting surface, using spin coating technique to form a thin film. The experimental results for single dye using masquerade tree leaf (A), pumpkin leaf (B) and pride of barbados flower (C) yield power conversion efficiencies (PCEs) of 0.45%, 0.72% and 1.21%, respectively. Also, the DSSCs based on blended dyes in a cocktail show that the PCE of 0.97%, 1.09% and 0.098% for masquerade tree leaf and pumpkin leaf (AB), masquerade tree leaf and pride of barbados flower (AC) and pumpkin leaf and pride of barbados flower (BC), respectively. The results show that pride of barbados flower (C) dye based DSSC has the highest PCE of 1.21% with an open-circuit voltage (V_{oc}) of 0.85 V, short-circuit current density (J_{sc}) of 3.59 mA/cm^2 and fill factor (FF) of 39.54% .

***Keywords*:** Dye-sensitized solar cells, Cocktail dye extract, Natural pigments, Anthocyanin dye extract, Chlorophyll dye extract

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Article Highlights

- Dye sensitized solar cells (DSSCs) were fabricated using single (pumpkin leaf, masquerade tree leaf and pride of barbados flower) and cocktail (blended) (masquerade tree leaf : pumpkin leaf, masquerade tree leaf:pride of barbados flower and pumpkin leaf : pride of barbados flower) dye extracts as active materials. The optical and current voltage (I-V) characterization were carried out.
- The DSSC scontaining single active materials of masquerade tree leaf, pumpkin leaf and pride of barbadosshow the power conversion efficiency of 0.45%, 0.72% and 1.21%, respectively.
- The DSSC containing cocktail dye extract of pumpkin leaf : pride of barbadosshows the least power conversion efficiency of 0.098%. This could be due to presence of reduced anchored-bonds between the cocktail dye extracts and TiO_2 molecules through which electrons transport from the excited dye molecules to the TiO_2 film.

1. INTRODUCTION

The pursuit of clean and sustainable energy sources to meet the growing energy demand has become a critical endeavor for humanity [1-4]. Among these sources, solar energy stands out as a renewable option that poses no threat to the public and can be utilized limitlessly. Dye Sensitized Solar Cells (DSSCs) serve as devices converting sunlight into electricity through the use of dyes as active materials [5, 6]. Within DSSCs, components such as the photoanode, active material (absorbing layer), electrolyte, and counter electrode play crucial roles [7].

The active material, or dye, absorbs light passing through the photoanode, causing electrons to move from the highest occupied molecular orbital (HOMO) to the lowest unoccupied molecular orbital (LUMO) of the dye. Subsequently, excited electrons are injected into the photoanode (TiO_2), transported to an external circuit, and directed to the counter electrode. The counter electrode then transfers electrons to an intermediate called the electrolyte, which restores the lost electrons to the photoanode [8]. While the most efficient DSSCs currently utilize synthetic Ruthenium (Ru) dye absorbed onto nanoscale TiO_2, achieving a power conversion efficiency (PCE) of 11-12%, the use of expensive metals like N3 and N719 presents certain drawbacks [9, 10]. Different inorganic, organic, and cocktail (hybrid) dyes have been used as photosensitizers [9, 11-14]. Due to expensive and toxic nature of synthetic dyes, various natural dyes have been used in DSSCs [13-17].

Chang *et al.*used chlorophyll and anthocyanin dyes from Wormwood and Red cabbage as photo-sensitizers in DSSCs and the power conversion efficiency reported are 0.90 % and 1.47 %, respectively [18]. Mamidi *et al.*used dried palash flowers, dried Hibiscus flowers, beet, pomegranate, red wine, mechandi, turmeric as active materials in DSSCs. They reported that dried hibiscus leaves as active material gave best result when deployed in DSSCs [19].Wongcharee *et al.* used blended rosella and blue pea pigments dyes in equal proportions to determine the performance of the blended dye, which achieved a maximum PCE of 0.37% [20]. Thambidurai *et al.*, fabricated DSSCs based on Ixora coccinea, Mulberry, and beetroot extract sensitized ZnO nanorod and reported that the PCEs are 0.33, 0.41, and 0.28%, respectively [21].

Göde, F *et al.*, investigated natural dye sensitizers, including Geneus Techtona grandis, Chassalia curviflora, and Lawsonia inermis, to enhance Dye-sensitized solar cells (DSSCs).

Co-sensitization and solvent effects (ethanol and deionised water) are examined. Tectona grandis in ethanol achieves the highest efficiency at 0.7%,attributed to its anthocyanin dervative's superior sensitivity to visbile light [33]. Prakash *et al.,* constructed DSSCs using sensitizers extracted from Amaranthus cruentus leaves and flowers,both fresh and dried. Using solvent facilitated dye extraction. A set of nine dyes underwent UV–visible and FTIR spectroscopy to confirm their suitability as DSSC sensitizers. The TiO_2 paste was characterized through XRD and SEM analysis. Nine DSSCs were assembled, incorporating TiO_2 as the semiconductor oxide, I^-/I^{3-} as the electrolyte, and graphene-coated FTO as the counter electrode. The highest efficiency of 0.816% for the cell using acetone-extracted dye from fresh leaves, outperforming previous studies using Amaranthus red dyes [34].

Park *et al*., deployed yellow Gardenia as natural photosensitizer and reported maximum power conversion efficiency of 0.35% [22]. S. C Ezike *et al*., presents fabricated DSSCs based on single and mixed dyes from Talinum fruticosum and Telfairia occidentalis (chlorophyll pigments) and Caesalpinia pulcherrima (anthocyanin pigment) and cyclic voltametry test shows that the dye are efficient solar absorbers. The results show that DSSCs based on mixed dye extract from Caesalpinia pulcherrima flower and Telfairia occidentalis at different mix ratios have the highest photovoltaic performance due to low charge transfer resistance and broader photo-absorption in visible region compared to both the single and other mixed dye extracts with optimized efficiency of 1.14%. However, the DSSCs based on mixed dye extracts from Talinum fruticosum and Telfairia occidentalis have the least photovoltaic performance among all the devices with efficiency of 0.13%. [36]. Also, Cho *et al*. Used cocktail of dyes extracted from four different kinds of vegetable dyes to fabricate DSSCs and they achieved PCE up to 1.57% [12].

This study uses single and cocktail (hybrid) of dyes, as sensitizers for DSSCs, extracted from three different kinds of plant dyes in order to reduce the cost of fabrication. The optical and photovoltaic properties of the different plant dyes extracts and DSSCs, respectively are determined.

2. MATERIALS AND METHODS

2. 1. Materials

The dye extracts were obtained from chlorophyllous plants: pumpkin leave (*Telfairia occidentalis*) and masquerade tree leaf (*Polyalthia longifolia*), and anthocyanin – containing pride of barbados flower (*Caesalpinia pulcherrima*) (Figure 1) in Hong, Hong local government area of Adamawa state, Nigeria. Fluorine doped tin oxide (FTO) substrate purchased from Dyesol was cut to size of 4 cm × 4 cm. The photoanode was TiO_2 nanoparticle purchased from Sigma Aldrich, electrolyte solution was purchased from EL-HPE Dyesol. TiO_2 is better photoelectrode as compare to ZnO [35]. HB graphite pencil used as carbon for counter electrode and Whatman filter paper was used to remove residues from anthocyanin and chlorophyll extracts.

2. 2. Substrate cleaning

FTO substrates were cleaned as reported elsewhere [23]. The substrates were immersed in distilled water with detergent in an ultrasonic bath (07043-986 Symphony) for 15 minutes at 27 °C. The detergent-cleaned FTO was cleaned with distilled water in ultrasonic cleaner for 15 minutes at 27 ℃. Later it was cleaned with ethanol and lastly, with Isopropyl alcohol at the same conditions and dried under nitrogen gas.

Figure 1. Sources of dye extracts (a) Masquerade tree leaf (*Polyalthia longifolia)* (b) Pumpkin leaf (*Telfairia occidentalis*) and (c) Pride of barbados flower *(Caesalpinia pulcherrima)*

2. 3. Preparation of photoanode and counter electrode

The TiO_2 pastes was prepared using TiO_2 powder by dissolving it in ethanol solution. The TiO_2 solution was then stirred by 300 rpm for 30 minutes using a magnetic stirrer and it was warmed up to a temperature of 60 °C using hot plate and the solution were allow to cool down to room temperature. TiO_2 solution was spin coated at 4000 rpm for 30 seconds on the FTO substrate and annealed in muffle furnace for an hour at 450 °C Counter electrodes were prepared from carbon catalyst on the conductive side of the FTO glass. Carbon black electrodes were easily made by moving the FTO conductive glass substrates (conductive side of the FTO) over the flame of candle soot until the conductive side of the FTO turns black, it was allow to cool down. Both prepared photoanode and counter electrodes were kept for further use.

2. 4. Dye extraction

The leaves and the flowers were cut into smaller sizes using a sterilized knife washed with ethanol and distilled water solution. The smaller-size leaves were air-dried for thirty (30) days under room temperature. The dried leaves and flowers were then grounded into powdered form with electric blender (Philips HR-2818). 10 g of the leaves and flower powders were measured using Ohaus weighing balance (HT-A1000) and dispersed in 100 ml of ethanol and left under dark room temperature for 18 hours. Thereafter, each of the dispersed was warmed up for 5 min on a hotplate at temperature of 78.37 °C with a continuous stirring using a magnetic stirrer at 1000 rpm. After cooling down to room temperature, the dispersions were filtered with Whatman filter paper (Cat No 1001 150) and kept in a beakers. As reported by Asful *et al.* [24] ethanol was used as a solvent because it has an ethyl group which is non-polar and polar hydroxyl to dissolving pigments.

2. 5. Solar cells fabrication

The TiO_2 electrodes were immersed into the various dye solutions for 18 hours. The dye solutions were covered with aluminium foil in other to block out light rays from degrading the dye solutions and to prevent evaporation. The adsorbed dye on TiO_2 deposited FTO substrates were removed and rinsed in ethanol solution, in order to remove excess dye. DSSCs were assembled following the schematic diagram described in Figure 2, the catalyst- coated counter electrode placed on the top so that the conductive side of the counter electrode faces the TiO_2 film. The electrolyte solution (Iodide/triiodide) was placed at the edges of the plates. The liquid electrolyte was drawn into the space between the electrodes by capillary action. Binder clips were used to hold the electrodes together.

Figure 2. Schematic diagrams of DSSCs arrangement

2. 6. Characterization and measurement

The absorbance of dyes solutions was characterized using UV-Vis spectrophotometer (JENWAY 6705). The I-V measurement wascarried using I-V source meter (Keithley 2400). Calculations and graphs plotted were carried out using Origin pro8 software.

3. RESULTS AND DISCUSSION

3. 1. Optical Absorption of Dye Extracts

The extracted dyes were characterized using UV-Vis spectrophotometer. The dye extracts (samples) were labeled using letters A, B and C for *Polyalthia longifolia*, *Telfairia occidentalis* and *Caesalpinia Pulcherrima*, respectively. Light absorption analysis in the dyes was conducted to determine the ability of the dye to capture visible light (400 to 800 nm). The optical absorption of the dyes is shown in Figure 3. The graph shows that each single dye contained pigments that have the various capabilities to absorb energy based on a certain range of wavelength. Figure 3 shows the absorption spectra for three natural dyes. The figure shows that the maximum absorption peak (absorption edge) for A is at 538 nm with absorption wavelength range of 523 ~ 556 nm. Sample A with maximum absorption wavelength of 538 has corresponding band gap energy of 2.3 eV. The maximum absorption peak for B is at 537 nm with corresponding absorption wavelength range of 521 ~ 559 nm. Sample B with maximum absorption wavelength of 537 also has corresponding band gap energy of 2.3 eV. Sample C has the maximum absorption peak at 666 nm, and its range of the absorption wavelength is 643 ~683 nm. Its maximum absorption wavelength of 666 nm has corresponding band gap energy of 1.9 eV. The band gap energy was calculated from the wavelength at the absorption edge of the dye extract [24]. The absorption spectrum of dye extract represents the minimum energy required for electronic transition from the valence band or highest occupied molecular orbital (HOMO) level to the conduction band or lowest unoccupied molecular orbital (LUMO) level [25].

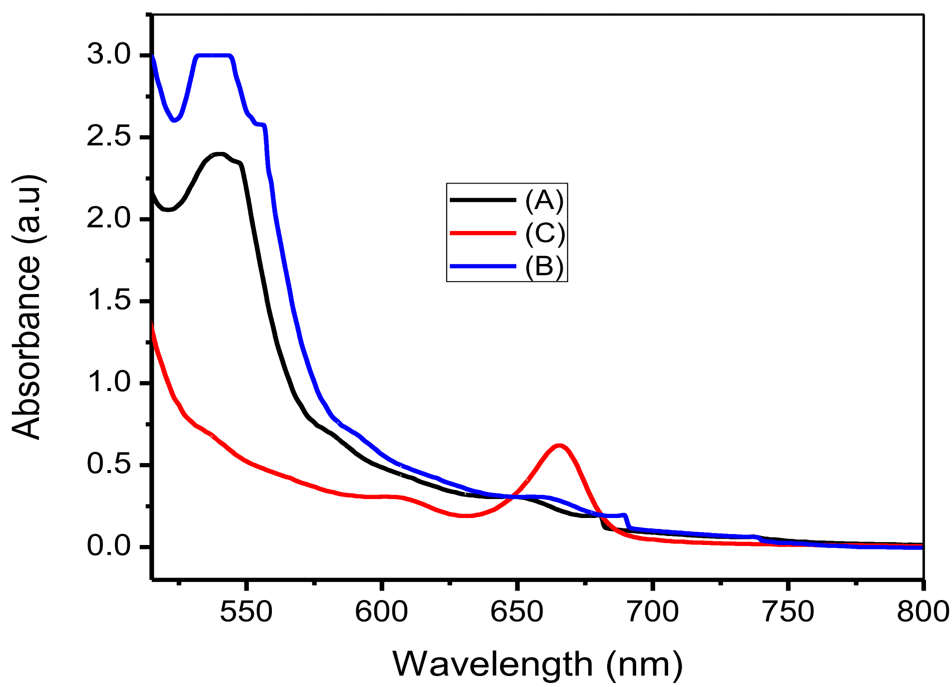

Figure 3. Absorption spectra of the extracted dyes from masquerade tree leaf (A) Pumpkin leaf (B) and Pride of barbados flower (C)

Figure 4 shows the absorption spectra for three types of cocktail (mixed) dyes labeled AB, AC and BC for masquerade tree leaf and pumpkin leaf, masquerade tree leaf and pride of Barbados flower, and pumpkin leaf and pride of Barbados, respectively. The purpose of cocktail dye is for the dye to absorb wider spectrum of solar radiation spectrum. Mixtures of several dyes with various absorption spectra are often prepared to obtain the maximum absorption within the visible and infrared region. For DSSCs applications, these mixtures co-sensitize the device to increase the global absorption using the widest wavelength range possible and thus maximizing the efficiency[17].Sample AB has absorption wavelength range of 521 ~ 572 nm and sample AC has absorption wavelength range of 523 ~ 524 nm and 598 ~ 638 nm. In addition, sample BC has absorption wavelength ranges of 521 ~ 548 and 584 ~ 625 nm. It shows that there is increase in absorption range wavelength of chlorophyllous cocktail dye extract and allows further absorption of more visible light as also reported by Cho *et al.*[12], which increases the photoelectric conversion efficiency of a DSSSC. Cho *et al*. [12] also reported broader absorption band in mixed sweet potato leaf and blueberry extracts in a volumetric proportion of 1:1. The cocktail extracts containing anthocyanin and chlorophyllous dyes show reduction in absorption ranges as compared to the respective absorption range of the single dye solutions. Reduction in absorption range of the cocktail extracts (B and C) could lead to lower performance of the cells when to compared to the single state of the dyes in DSSCs. Therefore, the broader absorption band and the good matching of HOMO/LUMO levels of the dye with the bottom edge of conduction band of semiconductor and the redox potential of electrolyte give the high efficiency of DSSC [25].

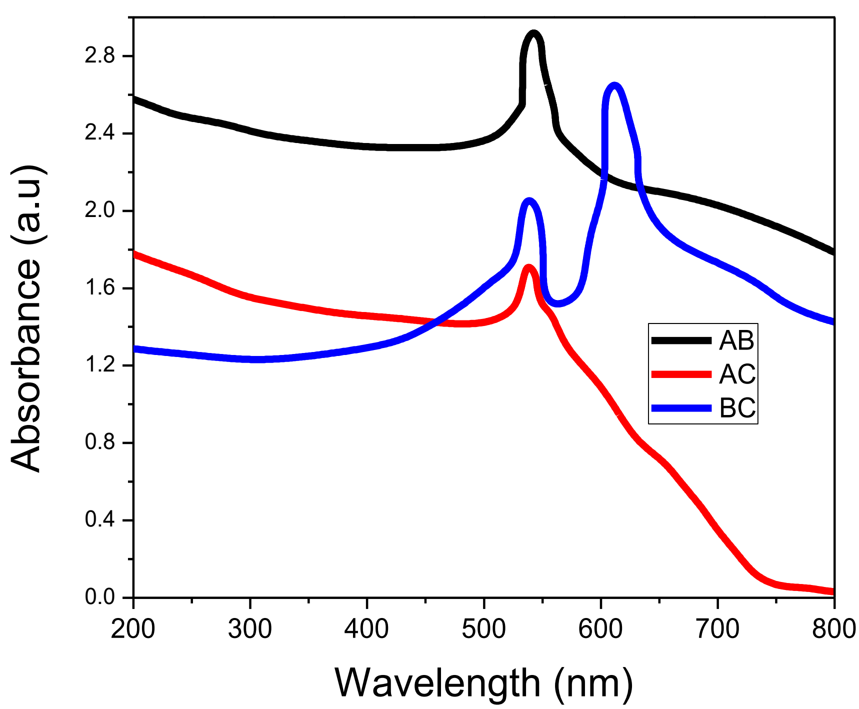

Figure 4. Absorption spectra of cocktail dyes from masquerade tree leaf and pumpkin leaf (AB), masquerade tree leaf and pride of barbados (AC), and pumpkin leaf and pride of barbados (BC)

3. 2. Adsorption of dyes on TiO_2

The dye-unloaded on TiO_2 sample absorbs in ultraviolet region while the dye-loaded on TiO_2 samples absorb at visible region (Figure 5). It is observed that dye-loaded on TiO_2 exhibit red-shift in the wavelength absorption maxima. It is expected that wavelength absorption maxima change is attributed to delocalization of electrons in the region. This suggests strong interaction between dye molecules and TiO_2 surface due to packed molecular conformation of dyes on TiO_2 matrix [26].The adsorption of dye on TiO_2 surface could be attributed to excellent adsorption ability of hydroxyl and carboxylate groups in the chlorophyllous and anthocyanin plant, respectively. The hydroxyl group adsorbs on the surface of the porous TiO_2 electrode while carboxylate group adsorbs at the defect sites of the TiO_2 [27].

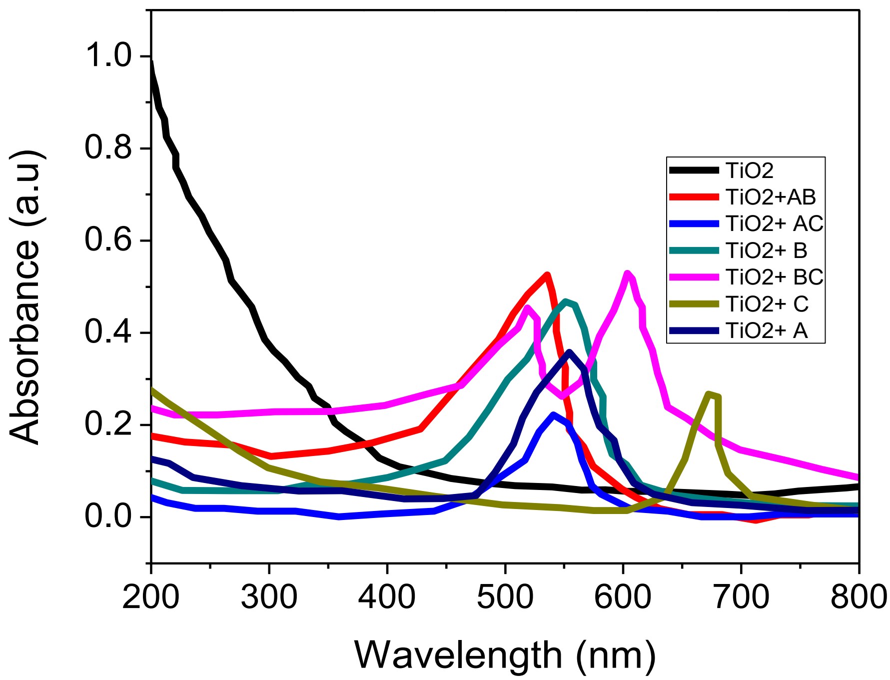

Figure 5. Optical adsorption of dyes on TiO2 film.

3. 3. Photovoltaic Properties of fabricated Cells

The photoelectric characterization of the fabricated cells (Figure 6) was examined under AM1.5 solar irradiation.

Figure 6. The fabicated DSSCs based on single and cocktail dye extracts

The current density – voltage (J-V) characteristics of DSSCs are depicted in Figure 7 and the device parameters are listed in Table 1. The DSSCs parameters were calculated using Equations(1) and (2) [28].

$$FF = \frac{P_{max}}{J_{sc} \cdot V_{oc}} \tag{1}$$

$$\eta = \frac{V_{oc} \cdot J_{sc} \cdot FF}{P_{light}} \tag{2}$$

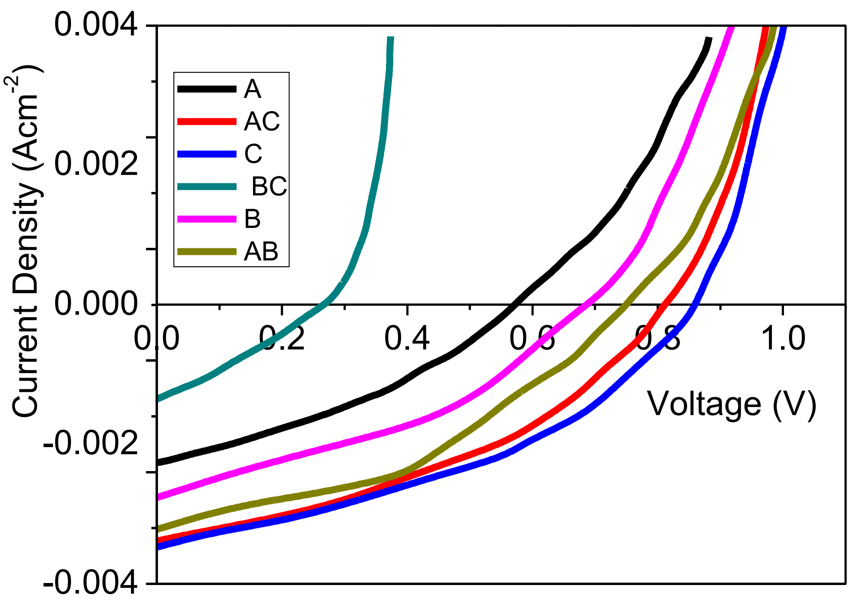

Figure 7. J-V plot of DSSCs fabricated from single and cocktail dye extracts

The pride of barbados flower, pumpkin leaf and masquerade tree leaf based single dye extracts have PCE of 1.21%, 0.72% and 0.45%, respectively (Table 1). The anthocyanin (pride of barbados flower) based DSSCs shows the highest PCE when compared with other chlorophyllous based DSSCs.

Table 1. Photoelectric parameters of single and cocktail dye extracts of the fabricated DSSCs

S/N	DSSC based dye	Symbol	J_{sc} (mAcm^{-2})	V_{oc} (V)	FF (%)	η (%)
1	Masquerade tree leaf	A	2.25	0.57	35.48	0.45
2	Pumpkin leaf	B	2.78	0.67	38.48	0.72
3	Pride of barbados flower	C	3.59	0.85	39.54	1.21

4	Masquerade tree leaf : pumpkin leaf	AB	3.24	0.74	40.46	0.97
5	Masquerade tree leaf : pride of barbados flower	AC	3.34	0.83	39.70	1.09
6	Pumpkin leaf : pride of barbados flower	BC	1.30	0.28	27.26	0.098

It is thus interesting to know why one, among the single dyes, outperforms the others. Examining carefully the device parameters for each group of DSSCs, higher PCE is mainly attributed to increase inV_{oc} of the device. It is expected that increase in V_{oc} in the device could lead to lower band gap energy of sensitizer dye which harvests photons of lower energies [29]. Also, it could lead to better electrons injection towards conduction band of TiO_2 due to reduced loss of photogenerated electrons [30]. Finally, increase in V_{oc} could lead to better dye-adsorption which would result in an increased dye coverage and reduced electron trapping at dye/TiO_2 interfacesd due to reduced defect state [31].

The cocktail dyes containing chlorphyllous extracts based DSSC shows enhancement in the photovoltaic performance of the cell. The enhanced performance is attributed to increase in V_{oc} and FF compared to the respective performances of the single dyes based DSSCs. The fabricated DSSC shows improved PCE of 0.97%. Meanwhile, the cocktail dyes containing chlorophyllous and anthocyanin extracts based DSSCs (AC and BC) have reduced PCE compared to the anthocyanin single dye based DSSC. The PCEs of fabricated cells AC and BC are 1.09% and 0.098% .

From the results obtained, it shows that there could be enhancedelectron transfer mechanism to the semiconductor electrode, dissociation process and efficiency of the cell in the blended dye extracts of chlorophyll-based DSSC. Meanwhile, mixture of chlorophyll and anthocyanin (Pumpkin leaf : pride of barbados flower) based DSSCs could reduce the electron transfer mechanism to the semiconductor electrode and dissociation process which leads to decrease in the cells efficiency. This could be due to presence of reduced anchored-bonds between the cocktail dye extracts and TiO_2 molecules through which electrons transport from the excited dye molecules to the TiO_2 film. This is influenced by the few of the raised exciton and the low amount of charge that reaches the electrode when the cell is irradiated by low of intensity light. The few of charge reaches the electrode, caused by the length of the charge diffusion path so that many charges are recombined before it reaches the electrode [32].

The photoelectric property of the fabricated dyes corresponds with the optical property of the dye extracts and agrees with related report on mixed dye in DSSCs [14].

4. CONCLUSIONS

In conclusion, the research delves into the development of Dye Sensitized Solar Cells (DSSCs) with a specific focus on the utilization of natural dye extracts,including chlorophyll and anthocyanin. The study explores the DSSCs' performance when employing these dyes individually and in combination, shedding light on critical aspects of photovoltaic efficiency. It reveals a nuanced relationship between the choice of dye extracts and their impact on DSSC performance. The work presents fabricated DSSCs from chlorophyll and anthocyanin dye extracts. The DSSCs are based on single and cocktail dye extracts. The results show that DSSCs based on cocktail extracts from anthocyanin (pride of barbados flower) and chlorophyllous (masquerade tree leaf and pumpkin leaf) plants have reduced photovoltaic performance due to narrower photo absorption in visible region compared to the single dye extract of pride of barbados flower. Meanwhile, the DSSCs based on cocktail extracts of chlorophyllous plants has enhaced Voc and FF which led to better photovolatic performance of the cell than the single dye based DSSCs. Therefore, the research underscores the pivotal role of the choice of sensitizers in DSSC performance. Cocktail extracts from chlorophyllous plants have demonstrated the potential to enhance the photovoltaic capabilities of these cells. In essence, this research demonstrates the complex interplay between dye sensitizers and DSSC performance, and the potential for more efficient and environmentally friendly energy generation.

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