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Photoelectric Characterization of a Dye-sensitized Solar Cell Based on Natural Pigment Extracted from Roselle (*Hibiscus sabdariffa*) Flower and TiO_2 Nanoparticles

Eli Danladi[1*], M. Y. Onimisi[1], S. G. Abdu[2], Ezeoke Jonathan[1] and S. O. Yakubu[3]

[1]Department of Physics, Nigerian Defence Academy, Kaduna, Nigeria.
[2]Department of Physics, Kaduna State University, Kaduna, Nigeria.
[3]Department of Mechanical Engineering, Nigerian Defence Academy, Kaduna, Nigeria.

Authors' contributions

This work was carried out in collaboration between all authors. Authors ED and MYO designed the study, undertook the experimental work, performed the statistical analysis, wrote the protocol, wrote the first draft of the manuscript and managed literature searches. Authors SGA, EJ and SOY managed the analyses of the study and literature searches. All authors read and approved the final manuscript

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ABSTRACT

Dye-sensitized solar cell (DSSC) was fabricated using natural dye extracted from Roselle flower. The sensitization performance related to interaction between the dye and titanium dioxide (TiO_2) surface was evaluated under 100 $mWcm^{-2}$ light intensity. The DSSC based on the *Hibiscus sabdariffa* organic dye gave a short-circuit current density (J_{sc}) of 0.0259 $mAcm^{-2}$, open-circuit voltage (V_{oc}) of 0.432 V and fill factor (*FF*) of 0.594, yielding an overall solar conversion efficiency (η) of 0.0067%. The result obtained shows some improvement over similar studies carried out by Adenike et al. [1] with lower efficiency of 0.002%. The improved performance might be attributed to the differences in concentrations of phytoconstituents in different parts of the plant and also the type of solvent used to extract the sensitizer.

**Corresponding author: E-mail: danladielibako@gmail.com;*

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1. INTRODUCTION

The most well-known and studied unconventional photovoltaic system is the dye- sensitized nanostructured solar cells. Incorporation of dye molecules in some wide bandgap semiconductor electrodes was a key factor in developing photoelecrochemical solar cells. Michael Gratzel and coworkers at the Ecole Polytechnique Federale de Lausanne [2,3,4] succeeded for the first time to produce what is known as "Gratzel Cell" or the dye sensitized solar cell (DSSC) to imitate photosynthesis, the natural processes plants convert sunlight into energy by sensitizing a nanocrystalline TiO_2 film using novel ruthenium (Ru) bipyridl complex. In dye sensitized solar cell, charge separation is accomplished by kinetics competition like in photosynthesis leading to photovoltaic action. The organic dye monolayer in the photoelectrochemical or dye sensitized solar cell replaces light absorbing pigments (chlorophylls), the wide bandgap nanostructured semiconductor layer replaces oxidized dihydro-nicotinamide-adenine-dinucleotide phosphate (NADPH), and carbon dioxide acts as the electron acceptor. Moreover, the electrolyte replaces the water while oxygen as the electron donor and oxidation product, respectively [5,6]. It has been shown that DSSCs are promising class of low cost and moderate efficiency solar cells based on organic materials [7,8].

DSSCs are based on the photo-injection of electrons from dye molecules into an inorganic semiconductor and holes transport by a redox mediator [9]. The nanoporous structure of the inorganic semiconductor provides the large surface area necessary to achieve significant optical density of the solar cell despite the low light absorption of a dye monolayer [10,11,12]. However, the porous electrode also plays an important role in the enhancement of recombination processes in the DSSC, thus decreasing all cell parameters and its total conversion efficiency [13,14,15]. Since the electrolyte penetrates throughout the entire porous structure, a large surface area is available for a reaction between the photo-injected electrons in the semiconductor and the oxidized ions in the redox mediator or oxidized dye at the semiconductor surface [13,16].

Major advantages of DSSCs are the large flexibility in shape, colour, transparency, and performance also under diffuse light. DSSCs could be integrated into large varieties of products, e.g. hand bags or clothing, indoor applications, and building-integrated photovoltaics such as rollable devices for walls of buildings or windows. But many components of a DSSC remain to be optimized. We focus on the identification and usage of natural pigments as light harvesting elements in DSSCs. Natural pigments as photosensitizers in DSSCs have the potential to reach similar performances and stability as known from dyes based on metal complexes.

In this work, we reported the performance of a DSSC sensitized with *hibiscus sabdariffa* with water as extracting solvent for the dye. The performance of this cell was compared with a similar cell fabricated under the same condition but with ethanol as extracting solvent. We found that, the cell fabricated with aqueous extract outperformed the DSSC fabricated with ethanol extract.

The *Hibiscus sabdariffa* (commercially known as Roselle) belongs to the family *Malvacae* and is present in abundance through-out the world and has attained prominence as a jute substitute [17].

1.1 Photovoltaic Performance

The dye used in dye-sensitized solar cells is extremely efficient at converting absorbed photons into free electrons in the titanium oxide TiO_2 layer. However, the current is limited to how many photons the dye can actually absorb. The photons that do get absorbed are the ones that ultimately produce the current. The rate at which the photons are absorbed depends on the overlap between the absorption spectrum of the titanium oxide layer (or other nanocrystalline oxide film used) and the solar flux spectrum. The maximum possible photocurrent is dependent on the overlap between these two spectra [18].

2. EXPERIMENTAL PROCEDURE

2.1 Dye Extraction

The flowers of *Hibiscus* were air dried till they became invariant in weight. The dried flowers of *Hibiscus sabdariffa* were left uncrushed because previous attempts proved failure to extract the dye from crushed samples due to jellification [19]. The method of heating in water was used to

extract the dye. Distilled water was the solvent for aqueous extraction. 5 g of the sample (Dried *Hibiscus sabdariffa* Flowers) was measured using analytical scale and dipped in 50 ml of the solvent heated to 100°C for 30 min after which solid residues were filtered out to obtain clear dye solutions. Roselle flower is riched in anthocyanins. It was reported that anthocyanin obtained from Roselle are delphinidin and cyanidin complexes [20]. The chemical structure of cyanidin and delphinidin in the *hibiscus sabdariffa* dye is shown in Fig. 4.

2.2 Preparation of TiO_2 Paste

The TiO_2 films were prepared using a modified sol–gel method, in which 2 g of P25 TiO_2 (Degussa, Japan) powder was dissolved in 10 ml of deionized water mixed with 0.2 ml of Triton-X 100 and 0.4 ml of acetaldehyde, then vibrated ultrasonically for 24 hours.

2.3 DSSC Assembly

The fluorine doped tin oxide (FTO) electrodes were washed with laury sulphate then later rinsed with water six times in an ultrasonication bath for ten (10) mins, then finally washed in propanol. The photoanode was prepared by first depositing a blocking layer on the FTO glass, followed by the nanocrystalline TiO_2. The blocking layer was deposited from a 2.5 wt% TiO_2 precursor and was applied to the FTO glass substrate by spin coating and subsequently sintered at 500°C for 45 mins. The 9 μm thick nanocrystalline TiO_2 layer was deposited by screen printing. It was then sintered in air for 30 mins at 500°C. The counter electrode was prepared by screen printing a platinum catalyst gel coating onto the FTO glass. It was then dried at 100°C and fired at 400°C for 30 mins.

The sintered photoanode was sensitized by immersion in the *hibiscus sabdariffa* extracts at room temperature overnight. The cell was assembled by pressing the photoanode against the platinum-coated counter electrode slightly offset to each other to enable electrical connection to the conductive side of the electrodes. Between the electrodes, a 50 μm space was retained using two layers of a thermostat hot melt sealing foil. Sealing was done by keeping the structure in a hot-pressed at 100°C for 1 min. The liquid electrolyte constituted by 50 mmols of iodide/tri-iodide in acetonitrile was introduced by capillary action into the cell gap through a channel previously fabricated at opposite sides of the hot melt adhesive. The channel was then sealed.

2.4 Characterization and Measurement

The current-voltage (I-V) data was obtained using a keithley 2400 source meter under AM1.5 (100 mw/cm^2) illumination and without illumination from a Newport A solar simulator, Scanning electron micrographs of the nanocrystalline TiO_2 films are taken with Carl Zeis SEM, The absorption spectrum of the dye was recorded on Ava-spec-2048 spectrophotometer, The cell active area was 1.8 cm^2. Thickness measurement was obtained with a Dektac 150 surface profiler. X-ray microanalysis was carried out with INCA EDX analyzer.

3. RESULTS AND DISCUSSION

Fig. 1a shows the SEM micrograph of TiO_2 film. From the figure, it shows that the TiO_2 nanoparticles produced have a mean particle size of about 18 nm. It also reveals that the surface is porous and has agglomeration.

Fig. 1b presents the EDX Image of TiO_2. The elements present in the TiO_2 are Titania, Chlorine, Oxygen and Nitrogen. Nitrogen is present due to the blower that was used to dry the TiO_2 semiconductor.

Fig. 2 shows the absorbance of the dye and TiO_2 with and without the dye within the wavelength range of 350-1000 nm. The absorption peak of the dye was observed around 550 nm (indicated in Fig. 2a), and the optical absorption enhancement was observed in the dye-loaded TiO_2 films. The relative changes in optical absorption of the TiO_2 film with and without dye are shown in Figs. 2b and 2c. The relatively broad and strong enhancement is observed in the range of 470–650 nm with a maximum peak around 550 nm, which coincides with the band position of decorated TiO_2 (Fig. 2c).

This enhanced absorption and broadened spectrum absorption of Fig. 2c was mainly attributed to the interaction between the dye and TiO_2. These features suggest that dye molecules in the environment of TiO_2 can absorb more photons, presumably due to the spectral overlap between the dye and TiO_2.

Fig. 3a shows the photocurrent density–voltage (*J-V*) curve of the DSSC under illumination and

Fig. 3b shows the *J-V* curve of the DSSC without illumination. In the dark, the cell behaves as a diode, and under illumination the *J-V* curve is shifted by the value of the current generated by the cell. Based on the *J-V* curve, the *fill factor* (*FF*) and *solar cell efficiency* (η) were determined using equations (1) and (2) respectively.

$$FF = \frac{P_{\max}}{P_{in}} = \frac{J_{\max} \times V_{\max}}{J_{SC} \times V_{OC}} \quad (1)$$

$$\eta = \frac{FF \times J_{SC} \times V_{OC}}{P_{IRRADIANCE}}.100\% \quad (2)$$

Where V_{max} = maximum voltage (V);
J_{max}=maximum current density (mA/cm^2);
J_{sc} = short current density (mA/cm^2);
V_{oc} = open circuit voltage (V) and
$P_{IRRADIANCE}$ = light intensity (mW/cm^2)

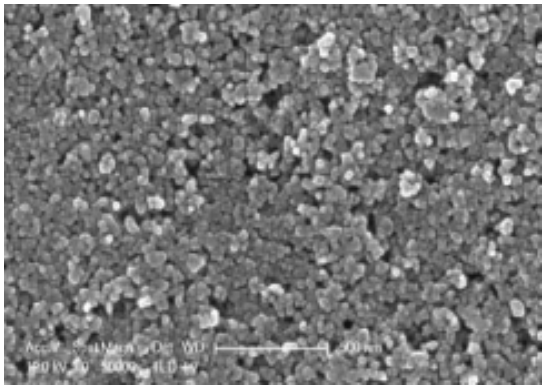


Fig. 1a. The scanning electron microscope surface morphology of TiO_2 sample

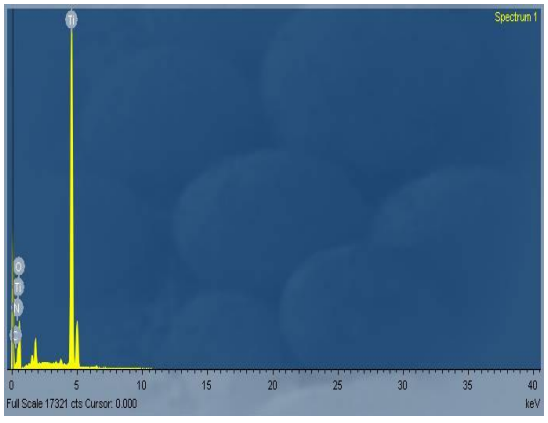


Fig. 1b. EDX image showing the elements present in the TiO_2 compound

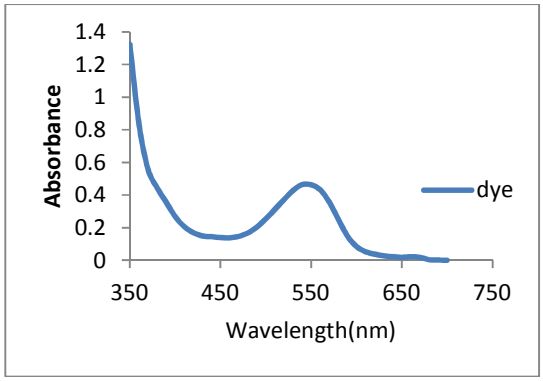


Fig. 2a. UV-vis spectra of the dye

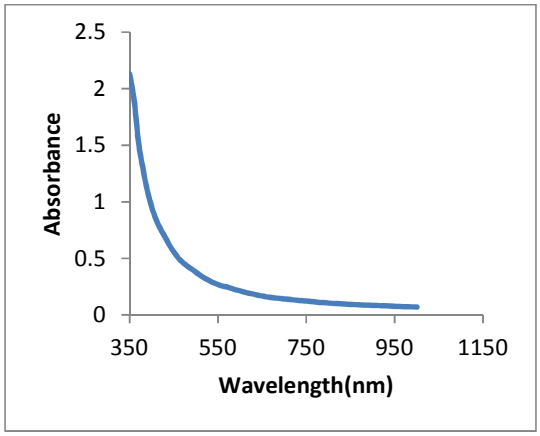


Fig. 2b. UV-vis spectra of TiO_2 without dye

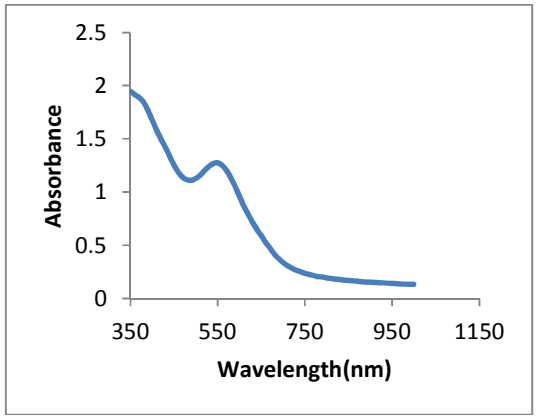


Fig. 2c. UV-vis spectra of TiO_2 with dye

The open-circuit voltage (V_{oc}), short-circuit current density (*Jsc*), fill factor (*FF*), and overall conversion efficiency (η) are summarized in Table 1. Adenike et al. [1] once reported an obtained solar energy conversion efficiency of 0.002 % using *hibiscus sabdariffa* extract with ethanol as extracting solvent. Better result was

obtained in our studies due to the differences in the extracting solvent. As shown in Fig. 3a and Table 1, the overall conversion efficiency was remarkably improved compared to the 0.002 % obtained in their result. This enhancement was caused by the used of distilled water as extracting solvent which allows better chemical connections between the dye and the semiconductor and subsequently lead to donation of more conduction electrons into the conduction band of the semiconductor instead of ethanol as extracting solvent as used in their research.

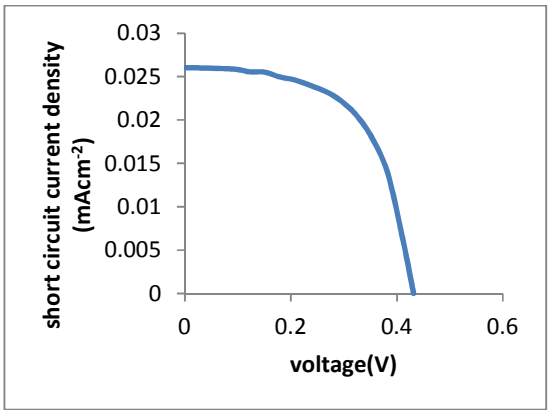


Fig. 3a. Photocurrent density-voltage (*J-V*) curve under illumination

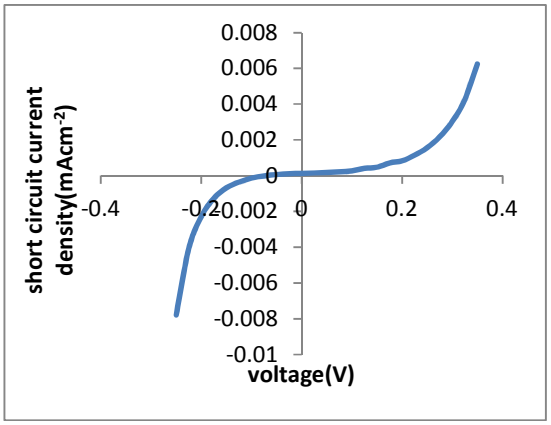
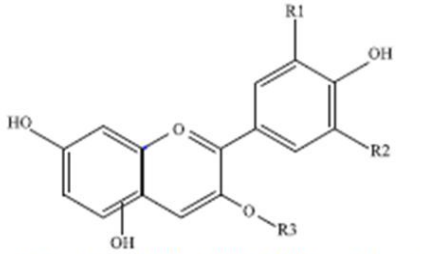


Fig. 3b. Photocurrent density-voltage (*J-V*) curve without illumination

Table 1. Performance characteristics of DSSC under 100 mWcm^{-2}

Sample	J_{sc}(mAcm^{-2})	V_{oc}(V)	FF	η(%)
DSSC	0.0259	0.432	0.594	0.0067



cyanidin-3-sambubioside (R1 = OH, R2 = H, R3 = sambubiose)
dephinidin-3-sambubioside (R1 = OH, R2 = OH, R3 = sambubiose)
cyanidin-3-glucoside (R1 = OH, R2 = H, R3 = glucose)

Fig. 4. Chemical structures of: Cyanidin and delphinidin in roselle dye

4. CONCLUSIONS

Dye sensitized solar cell was fabricated from anthocyanin extracts of *hibiscus sabdariffa* flower. The dye absorbs visible light in the range of 470 nm to 650 nm and its peak absorbance value was at 550 nm. The dye is chemically connected to a porous layer of a wide band-gap semiconductor. The DSSC employing the nanocrystalline TiO_2 as photoanode shows a power conversion efficiency of about 0.00670%. Poor charge transfer between the dye molecule and the TiO_2 due to low regeneration kinetics could be responsible for the low conversion efficiency of the cell [21]. The results obtained are encouraging and should prompt more detailed studies to uncover the exact mechanism involved.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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