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Dye-Sensitized Solar Cell (DSC) Based on Titania Nanoparticles and *Hibiscus sabdariffa*

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Authors' contributions

This work was carried out in collaboration between all authors. Authors TOA, POA and NA designed the study. Author TOA wrote the protocol and wrote the first draft while, authors NA and MBA carried out the experimental work and analyses. Authors TOA and MBA managed the literature searches. All authors read and approved the final manuscript.

Research Article

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ABSTRACT

Natural dye extracts from *Hibiscus sabdariffa* (popularly called Zobo in Nigeria; Western part of Africa) has been used as a sensitizer for nanosized titania (TiO_2) synthesized via hydrothermal route. The SEM-EDS analysis revealed the wide band gap titania (TiO_2) to be almost isomorphic ($\geq 99\%$ phase purity) with spherical particles having diameters in the range 25-40nm. The fabricated Dye-Sensitized Solar Cell (DSC) was tested and found to have photo generated current density of $0.17 mAcm^{-2}$ and open-circuit voltage value of 460 mV. The fill factor and power-conversion efficiency of the fabricated DSC were determined to be 41% and 0.033% respectively. This combination of natural dye extract with nanosized titania coupled with an appropriate hole transport material promises to provide a cheap, renewable and stable solar cells, paving way for large-scale commercialization in contrast to the expensive silicon based solar cells.

Keywords: Hibiscus sabdariffa; DSC; nanosized titania; photocurrent; open-circuit voltage; power-conversion efficiency.

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1. INTRODUCTION

Dye-Sensitized Solar Cell (DSC) has emerged as an environmentally friendly newer photovoltaic technology. It is associated with low cost of fabrication coupled with cheap component materials needing no high purity like the celebrated silicon-based solar cells. In the last two decades, extracts from *Hibiscus sabdariffa* popularly called Zobo almost took the place of “bottled drinks” in Nigeria because of its low cost and medicinal characteristics. With this emerging newer energy technology, *Hibiscus sabdariffa* (Zobo) may possess added economic value as a potential natural dye extract for the DSC, leading to an increase in the per capital income of Nigeria. DSCs are inimitable compared with almost all other kinds of solar cells in that electron transport, light absorption and hole transport are each dealt with by different materials in the cell [1,2]. The dye acting as a sensitizer in a DSC is coated onto the surface of a wide-band-gap semiconductor such as TiO_2, SnO_2 or ZnO, and then coupled to a platinized counter-electrode.

During the 1990s and the early 2000s, researchers found that organometallic complexes based on ruthenium provided the highest power-conversion efficiencies [3,4]. However, these organometallic complexes based on ruthenium contain intricate heavy rare-metals which are expensive to synthesize and not friendly to the environment [5]. This stringent effect of the complexes on the environment has paved way for natural dyes with acceptable efficient metal-to-ligand charge transfer and ease of extraction with low cost and are readily available in nature from various plants’ shoots and roots with colours raging from red to purple [6,7]. The possibility of using natural dye extracts containing anthocyanins and carotenoids for DSCs have been investigated and found to show different sensitizing performances [8,9]. In this work, we have demonstrated the fabrication of cost effective and stable *Hibiscus sabdariffa* Sensitized Solar Cell (HSSSC) with fill factor and power-conversion efficiency of 41% and 0.033% respectively. This combination of natural dye extract from *Hibiscus sabdariffa* with nanosized titania (TiO_2) coupled with an appropriate hole transport material promises to provide a cheap, renewable and stable solar cells, paving way for large-scale commercialization in contrast to the expensive silicon based solar cells in Nigeria and the world at large.

2. MATERIALS AND METHODS

Titanium (IV) chloride, ethanol, ammonium hydroxide and ammonium sulphate were obtained from BDH. Transparent conductive oxide coated glass [TCO, 15 ohm/m^2, $(1.00\times1.00)cm^2$], electrolyte (iodolyte-AN-50), Sealing gasket (surlyn-SX1170-25PF), and screen-printable platinum catalyst, (Pt-catalyst T/SP) all from SOLARONIX. Dye extract was obtained from the natural product *(Hibiscus sabdariffa).*

Titania nanoparticles were synthesized using titanium (IV) chloride via acid hydrolysis technique with ammonia solution as pH adjuster. A solution of 1.5M ammonium sulfate which consisted of 0.75M titanium (IV) chloride was used. The total volume of 50ml in aqueous solution was heated at 75ºC and monitored at the same temperature for 90 minutes. Subsequently, ammonium hydroxide solution (2.5M) was added drop-wise under high speed stirring until the pH reached 7.0. The precipitated titanium hydroxide was collected, repeatedly washed with distilled water and ethanol and dried at 50-55ºC. After being calcined at 400ºC for 4 hours, the sample was slowly cooled at room temperature. Dried leaves of *Hibiscus sabdariffa* were crushed into tiny bits and boiled in 75ml of deionized water for 15 minutes. The residue was removed by filtration and filtrate was used as prepared. The resulting extract was centrifuged to further remove any solid residue and was used directly as prepared for the construction of the DSCs at room temperature.

TiO_2-paste was screen printed on a pre-cleaned fluorine doped tin-oxide (FTO) conducting glass and allowed to dry for 6 minutes for 125°C in open air. The TiO_2/FTO glass electrode was sintered in a furnace at 450°C for 30 min and allowed to cool to room temperature to melt together the TiO_2 nanoparticles and to ensure its good mechanical cohesion on the glass surface. The TiO_2 electrodes thickness was determined by Dekker Profilometer to be 9µm. The electrode was immersed (face-up) in the natural dye extracts for 18h at room temperature for complete sensitizer uptake. This turned the TiO_2 film from pale white to sensitizer colour. The excess dye was washed away with anhydrous ethanol and dried in moisture free air and. A DSC of 0.66 cm^2 active area was assembled by sandwiching a surlyn polymer foil of 25 µm thickness, as spacer between the photoanode and the platinum counter electrode and then hot-pressed at 80°C for 15 s. A drop of liquid electrolyte was introduced into the cell assembly via a pre-drilled hole on the counter-electrode and sealed using amosil sealant. In order to have good electrical contacts, a strip of wires were attached to both sides of the FTO electrodes.

Surface morphology of the synthesized TiO_2 nanoparticles was observed using EVOI MA10 (ZEISS) multipurpose scanning electron microscope operating at 20kV employing secondary electron signals, with the corresponding Energy Dispersive Spectroscopy (EDS) spectra obtained using characteristic x-rays emitted by TiO_2 nanoparticles. UV-visible absorption measurements of the dye extract and the screen printed TiO_2 were carried out with Avante UV-VIS spectrophotometer, model-LD80K. Photo electrochemical measurements of DSC were performed under a standard solar radiation of 100 mW/cm^2 using overhead Veeco-viewpoint solar simulator (equipped with AM 1.5 filter) - model 4200SCS and a four point Keithley-Multimeter coupled with a Lab-tracer software was used for data acquisition at room temperature. Subsequently, the reversely directed currents and the corresponding voltages were acquired at room temperature with an active cell area of 0.66 cm^2.

Based on the I-V curve, power conversion efficiency (η) was calculated according to the equation:

$$\eta = \frac{\left(FF \times J_{SC} \times V_{OC}\right)}{I}$$

where J_{sc} is the short-circuit voltage (volts), I is the intensity of the incident light (W/m^2), V_{oc} is the open circuit voltage (volts), FF is the fill factor defined as:

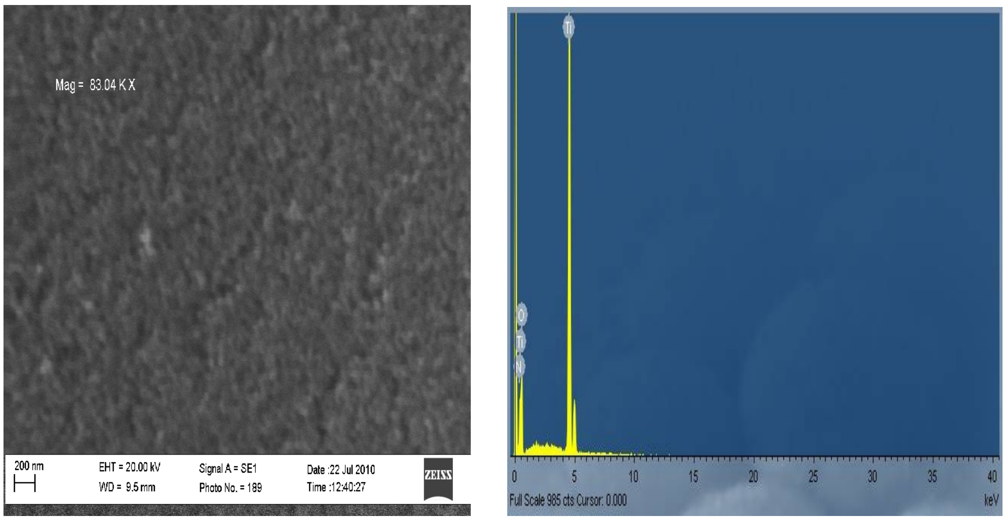
$$FF = J_m V_m / J_{SC} V_{OC}$$

where J_m and V_m are the optimum photocurrent and voltage that can be extracted from the maximum power point of the I-V characteristics [10].

3. RESULTS AND DISCUSSION

The image presented in Fig. 1. together with the corresponding EDS spectra obtained using characteristic x-rays emitted by TiO_2 nanoparticles was observed at a magnification of 83.04kX. The uniform contrast in the image revealed TiO_2 to be almost isomorphic.

Nevertheless, Oxygen and Nitrogen occur with minor concentrations as impurities thereby making Ti the dominant element with concentration of about 99.5% as depicted in the EDS spectra (Figure 1b). The morphology of TiO_2 nanoparticles is such that the particles are closely parked and spherical in shape. The average diameter of the particles is in the range of 25-40nm reflecting that TiO_2 nanoparticles are transparent and suitable for DSC application.



(a) **(b)**

Fig. 1. TiO_2 Powder Characteristics: (a) Surface Morphology of TiO_2 and (b) EDS spectra of TiO_2

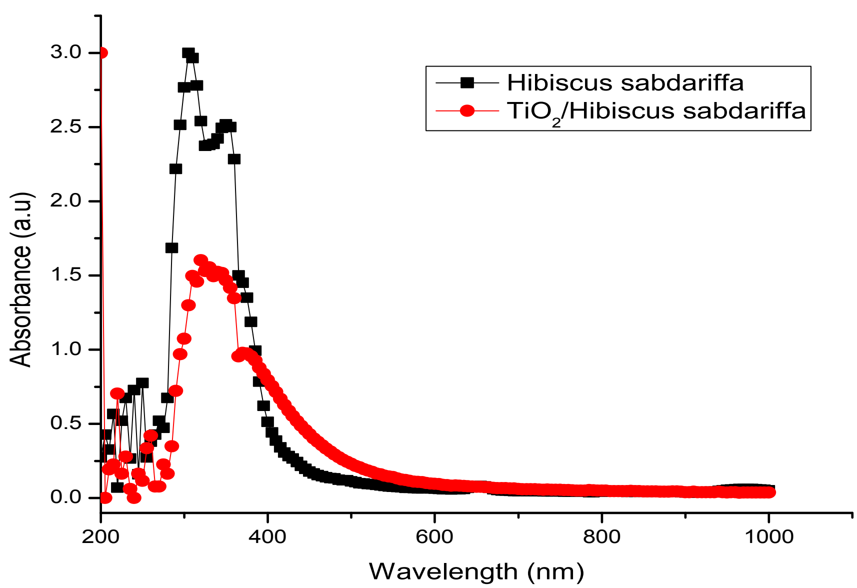


Fig. 2. UV–VIS absorption spectral of Hibiscus sabdariffa extract and TiO_2/*Hibiscus sabdariffa* extract

Fig. 2. shows the UV–VIS absorption spectral of extract and TiO_2. It can be seen that the dye extract exhibits a maximum at 310 and 380 nm, while TiO_2 /dye extract at 320 nm shows an absorption maximum (Fig. 2). In the TiO_2/dye extract, there are four shoulders as compared to the two shoulders in the dye extract and the band is broadened with a shoulder near 400nm.

Current density and power vs. voltage characteristics of DSC made with *Hibiscus sabdariffa* dye are plotted and shown in Fig. 3. The photovoltaic parameters are determined and tabulated in Table 1. The maximum current density is obtained to be 0.17mAcm^{-2}; the open circuit voltage and the fill factor are obtained to be 0.46V and 41% respectively.

Table 1. Photoelectrochemical parameters of the DSC sensitized with *Hibiscus sabdariffa*

DSC	J_{sc}(mAcm^{-2})	V_{oc}(V)	FF	η(%)
TiO_2/*H. sabdariffa*	0.17	0.46	0.41	0.033

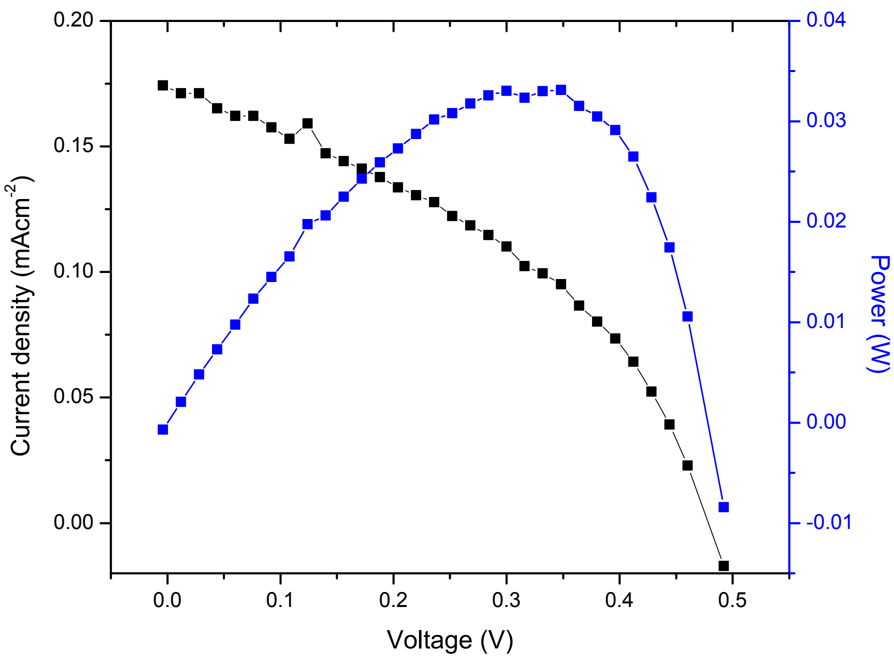


Fig. 3. Current density Power Vs voltage for DSC sensitized with *Hibiscus sabdariffa*

The power conversion efficiency of the cell sensitized with extracts from *Hibiscus sabdariffa* is 0.033%. This could be due both to possible electron/dye cation recombination pathways and sensitization at 320nm (as depicted in Fig 2). In fact, it is well known that proton adsorption causes a positive shift of the Fermi level of the TiO_2, thus limiting the maximum photovoltage that could be delivered by the cells [11]. Nevertheless, pure *Hibiscus sabdariffa* proved to be a rather poor sensitizer, as can be seen by the low spectral absorption at lower energies. However, no deviation from this trend was observed when the duration of immersion was varied. However, the cell shows no reduction in the efficiency under continuous stimulated sunlight illumination for several hours.

4. CONCLUSION

In this work we have reported an investigation on *Hibiscus sabdariffa* dye extracts as natural sensitizers of TiO_2 films for DSC. The best overall solar power conversion efficiency of 0.033% was obtained, under AM 1.5 irradiation and a maximum current density of 0.17mAcm^{-2}. Nevertheless, pure *Hibiscus sabdariffa* proved to be a rather poor sensitizer, as can be seen by the low spectral absorption at lower energies. Although the efficiencies obtained with this natural dye is still below the current requirements for large scale practical application, the results are encouraging and may boost additional studies oriented to the search of new natural sensitizers and to the optimization of solar cell components compatible with such dye. It has been pointed out that often raw natural dye mixtures exhibit better performance than commercial or purified analogues. This could be related to the presence in the natural extract of specific pools of ancillary molecules (*i.e.,* alcohols, organic acids, *etc.*) which act as coadsorbates, suppressing recombination with the electrolyte, reducing dye aggregation and favouring charge injection. In this light, we are currently exploring the possibility of increasing the power-conversion efficiency of DSC based on TiO_2 using raw natural dye mixture consisting of *Hibiscus sabdariffa* and *Butea monosperma*.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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