

Growth and Characterization of Dye-Sensitized Solar Cells using Dyes from Mangifera Indica, Manihot Esculenta and Hibiscus Sabdariffa Leaves by Sol-Gel Technique

MORKA, JC; OTTIH, IE; UMEOKWONNA, NS

Department of Industrial Physics, Chukwuemeka Odumegwu Ojukwu University, Uli, Anambra State, Nigeria

*Corresponding Author Editor: Johnmorka2@gmail.com Co-Authors Email: ie.ottih@coou.edu.ng; ns.umeokwonna@coou.edu.ng

ABSTRACT: Three natural dyes, extracted from leaves of mangifera indica, manihot esculenta and hibiscus sabdariffa were used as sensitizers to fabricate dye-sensitized solar cells. The Sensitization performance related to interaction between the dyes and titanium (iv) oxide (Ti0₂) surface were evaluated under 100mW/cm² light intensity. The photoelectrical performance of the DSSCs based on these dyes show that the V_{0c} ranged from 0.300 to 0.477V and J_{sc} was in the range of 0.00011 to 0.0102mA/cm². The DSSC sensitized with mangifera indica leave extract offered the best photosensitization effect, and had the highest photoelectrochemical performance. The SEM result TiO₂ sample revealed that the surface of the TiO₂ was porous with particle agglomeration. The EDX showed quantitative results and elements present as titania (70.33, weight %), oxygen (15.30, weight %), nitrogen (10.24, weight %) and carbon (4.13, weight %).

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Nowadays, the main method of utilization of solar energy is the conversion of solar energy into other energy sources. In 1954 the silicon solar cell developed by Bell marks that human can make solar energy converts into electrical energy for use, is epoch-making significance (Chapin et al; 1954). However, it is not suitable for large-scale usage, since this type of cell has the more stringent requirements for raw materials and production process. Though the development and deployment of amorphous and polysilicon solar cells for large-scale use subsequently followed, their simple production process notwithstanding, high costs still could not meet the large-scale use. In 1991, Professor Gratzel reported a new low-cost chemical solar cell by the successful combination of nanostructured electrodes and efficient charge injection dyes, known as Gratzel cells or dyesensitized solar cells which gave a photoelectric conversion efficiency of 7% under simulated sunlight irradiation (O'Regan and Gratzel (1991). It was designed to imitate photosynthesis; the natural

processes plants convert sunlight into energy by sensitizing a nanocrystalline TiO₂ 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-nicotinamideadeninedinucleotide phosphate (NADPH) and carbon (iv) oxide acts as the electron acceptor. In the same way, the electrolyte replaces the water while oxygen as the electron donor and oxidation product respectively. (Smestad and Gratzel (1998); Hara and Arakawa; 2003). It has been shown and demonstrated that DSSCs are promising class of low cost and moderate efficiency solar cells based on organic materials (Hagfeldt and Gratzel (1995). Haruna et al., (2015). The DSSC promises extremely cheap photovoltaic energy production by combining the advantages of non-vacuum processing, extremely low costs components, low embodied energy of production, potentially high efficiencies and superior performance compared to silicon solar cells under diffuse light conditions. In this paper, we extracted three natural dyes from leaves of *mangifera indica, manihot esculenta* and *hibiscus sabdariffa*; and these dyeswere used as sensitizers to fabricate dye-sensitized solar cells (DSSCs).

MATERIALS AND METHODS

Methods employed in extracting the dyes: The mangifera indica, manihot esculenta and hibiscus sabdariffa leaves were each collected from their trees respectively, washed and exposed to dry. mangifera indica, manihot esculenta and hibiscus sabdariffa leaves were air dried in a shade to prevent pigment degradation (Eli, et al, 2016). Each of the samples were grinded to small particles separately using ceramic mortar and pestle. 10g of Mangifera Indica, Leaves was mixed with 25ml of ethanol (99% absolute as the extracting solvent) to extract the dye. The mixture was then filtered and the filtrate was then stored in a test tube. Likewise, 10g of manihot esculenta and hibiscus sabdariffa leaves were mixed with 25ml of absolute ethanol to extract the dye. The mixture were then filtered and the filtrate were then stored in a test tube.

*Preparation of the TiO*₂ *paste:* The *TiO*₂*paste* was prepared by adding 5 mL (in 1 -ml, increments) of acetic acid solution (pH 4 in deionized water) 3g of colloidal *Degussa P25TiO*₂powder in a mortar and pestle while grinding. One drop of Triton X 100 (surfactant) was added to the mixture which will allow the final suspension to coat the glass substrates uniformly. The *TiO*₂ suspension (diluted white paste) was then stored in a dropper bottle.

DSSCs Assembling: All the materials were first cleaned and rinsed with distilled water distilled water and dried. The photoanode was prepared by first depositing a blocking layer on the FT0 glass (solarionix), followed by the nanocrystalline TiO₂. The blocking layer was deposited from a 2.5wt% Ti₀₂ precursor and was applied to the FTO glass substrate by spin coating and subsequently sintered at 400°C for 30 mins. The nanostructured TiO₂ layer was deposited by screen printing. It was then sintered in air for 30mins at 500°C. The counter electrode was prepared by using the candle flame carbon soot onto the FT0 glass. It was then dried at 100°C and fired at 400°C for 30mins. The sintered photanode was sensitized by immersion in the sensitizer solution at room temperature overnight. Sensitization was achieved by

immersing the photoanode in the extracts. The cells were assembled by pressing the photoanode against the carbon soot coated counter electrode slightly offset to each other to enable electrical connection to the conductive side of the electrodes. The photoanode and the carbon soot coated counter electrodes were assembled to form a DSSC by creating a gap of 50um between the two electrodes to be filled with 50mmol of iodide/tri-iodide dissolved in acetonitrile.

Characterization and Measurement: The absorption spectrum (light absorbance) of the chlorophyll (liquid extracts) were measured suing Ava-Spec-ULS 2048CL-2EV0 Spectrophotometer. The wave length range measured in this study was 300-1100nm. The scanning electron microscope (SEM) micrographs were taken with Phenom Pro X Model, Eindhoven de Netherlands SEM. Also the energy dispersive X-ray (EDX) characterization was carried out using the same equipment. The solar simulation was carried out using a Newport 94082A Solar Simulation machine and Agilent 8453 IET analyzer Current density and voltage (J-V) characteristics of the DSSCs were measured under simulated AM 1.5 sunlight at a light intensity of 100mW/cm². The effective irradiated area of each cell was 0.5cm².

RESULTS AND DISCUSSION

Operational processes of DSSC: Step 1: The dye molecule is initially in the ground state (D). The semiconductor material of the anode is at this energy level (near the valance band) non- conductive. When light shines on the cell, dye molecules get excited from their ground state to a higher energy state (D*), equation (1). The excited dye molecule has now a higher energy content and overcomes the band gap of the semiconductor.

$$TiO_2 - D + hv \rightarrow TiO_2 - D^*$$
 1
Dye excitation by photon

Step 2: The excited dye molecule (D*) is oxidized, equation (2) and an electron is injected into the conduction band of the semiconductor. Electrons can now move freely as the semiconductor is conductive at this energy level. Electrons are then transported to the current collector of the anode via diffusion processes (Smestad and Gratzel, 1998). An electrical load can be powered if connected. `

 $\text{TiO}_2-\text{D}^* \rightarrow \text{TiO}_2 + \text{D}^+ + e^-CB = 2$ Electron generation to CB of TiO₂ in ps scale

Step 3: The oxidized dye molecule (D^+) is again regenerated by electron donation from the iodide in the electrolyte (Hagfeldt and Gratzel, 1995) equation (3)

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 $TiO_2 - D^+ + 3I^- \rightarrow TiO_2 - D + I_3$ 3 Dye generation by electrolytic reduction in TiO2 in μs scale

Step 4: In return, iodide is regenerated by reduction of triiodide on the cathode (equation 4)

 $I_3 + 2e \rightarrow 3I^-$ 4 Reduction of electrolyte by the counter electrode



Fig 3: Absorbance spectra of mangifera indica, manihot esculenta and hibiscus sabdariffa leaves extract

Fig 3 shows the absorbance spectra of the dye extracted from *mangifera indica, manihot esculenta and hibiscus sabdariffa* leaves. There is an exponential decrease in absorbance with wavelength in the UV-VIS-NIR regions in the three leaves extract.

The highest absorbance values of 0.91 (19%), 0.64 (64%) and 0.32 (32%) at 300nm wavelength were exhibited by the *mangifera indica, manihot esculenta and hibiscus sabadariffa* leaves extract respectively.

The extract of *mangifera indica leaves and manihot esculenta* also in the figure, shows absorption peaks at 390nm and 380nm respectively. In the VIS region at 550nm wavelength, *mangifera indica, manihot esculenta and hibiscus sabdariffa* leaves had absorbance value of 0.16, 0.12 and 0.04 respectively. This is attributed to the presence of anthocyanins.

The chemical absorption of these dyes is expected to occur between of the formation of bond with the surface of nanostructured TiO_2

Current-voltage, (J-V) curves of the DSSCs characteristics for *mangifera indica, manihot* esculenta and hibiscus sabdariffa leaves extract showing the light and dark illumination respectively. From the curves, V_{oc} , J_{sc} , J_{max} , V_{max} were determined.

The FF and Photoelectrochemical performance efficiency were evaluated using equations (1) and (2) (Danladi *et al*; (2016) respectively.

$$FF = \frac{Pmax}{Pin} = \frac{Jmax.Vmax}{Jsc.Voc}$$
(1)

And

$$\eta = \frac{Jsc \times Voc \times FF}{Power_{irradiance}}$$

$$\eta = \frac{Jsc \times Voc \times FF}{A_c \times E}$$
(2)

Where; $V_{max} = maximum$ voltage (V); $J_{max} = maximum$ current density (mA/cm²); J_{sc} = short circuit current density (mA/cm²); V_{oc} = open circuit voltage (V) and $P_{irradiance} = Ac \times E$

Where: Ac = effective area of the cell in cm²; E = the input light in ^W/cm²; $\eta = IPCE$ = incident photon to current-conversion efficiency

Photovoltaic test of DSSCs using these natural dyes as sensitizers are summarized in Table 1.



Fig 4 shows the current-voltage, (J-V) curves of the DSSCs characteristics for mangifera indica, manihot esculenta and hibiscus sabdariffa leaves extract showing the light and dark illumination respectively.
Table 1: Efficiency Performance parameters of DSSCs with the three sensitizers under 100mW/cm²

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Sample	Leaves photo	J _{5C} (mA/cm ²)	Voc (v)	FF	Vmax (v)	Jmax (A/cm ²)	Ц (%)
Mangifera indica	X	0.0102	0.477	0.241	0.240	4.90 x 10 ⁻⁷	2.35 x 10 ⁻⁴
Manihot esculenta	*	0.00011	0.400	0.267	0.210	5.60 x 10 ⁻⁴	2.35 x 10 ⁻⁵
Hibiseus sabdariffa	-	0.000232	0.300	0.237	0.150	2.32 x 10"	6.97 x 10 ⁻³

As displayed in Table 1 and figure 4 and from the effective area of 0.5cm² the FF of these DSSCs as evaluated varied from 0.237 to 0.267. The Voc varied from 0.300 to 0.477V, and the J_{sc} changes from 0.000232 to 0.0102mA/cm². Specifically, a high Voc (0.477V) and $J_{sc}(0.0102mA/cm^2)$ were obtained from the DSSC sensitized with the mangifera indica leaves extract. The efficiency of the DSSC reached 0.000235%. The variations in the efficiencies is due to energy transfer mechanism where each dye molecule contain various anthocyanin pigments that absorb at certain wavelength (Motlan and Panggabean, (2020).





Fig 5 shows the SEM surface morphology of TiO₂ sample

From figure 5, it shows that the TiO₂ nanoparticles produced have a mean particle size of about 15nm. It also reveals that the surface of the TiO₂ is porous with particle agglomeration. Figure 6 presents the energy dispersive X-ray image of TiO2 EDX showed quantitative results and elements present as titania (70.33, weight %), oxygen (15.30, weight %), nitrogen (10.24, weight %) and carbon (4.13, weight %). Nitrogen is present probably due to the blower that was used to dry the TiO₂ semiconductor. This agrees with results obtained by (Danladi et al; (2016). The other elements seen were due to the FTO glass used. Same result has been reported by (Nwanya et al; (2012). It is clearly seen from the figure that Titania has the highest peak.

Conclusion: The performance of dye sensitizers' solar cells with three naturals dyes from leaves of mangifera indica, manihot esculentra and hibiscus sabdariffa was successfully investigated. Among the three dyes, the extract obtained from mangifera indica showed the best sensitization effect. The optical characterization of the three dyes revealed high absorbance in the visible (VIS) region of the spectrum, thus positioning them for photovoltaic solar cell device and fiber optic technology applications for solar energy harnessing and development

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Spectrum 1

40

keV

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