



Investigation of the Efficiency of Dye-Sensitized Solar Cell Using Natural Dyes as Photo Sensitizer

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Abstract: Dye-sensitized solar cells (DSSCs) were constructed by using the black plum, grapes skin, beetroot, mangosteen, and raspberry as natural sensitizers of anatase-based nanostructure TiO₂ thin film coated on fluorine-doped tin oxide (FTO) conducting glass. *N,N,N*-tris-(2-methoxy-naphthalene-1-yl)-*N,N,N*-triphenylbenzene-1,3,5-triamine [MNTT] was used as hole transporting material. The photovoltaic properties of the cell have been studied and the best overall solar energy conversion efficiency of 0.94% was obtained under air mass (AM) 1.5 irradiation.

Key Words: natural photosensitizers; DSSCs; solid state electrolyte; photovoltaic; solar energy.

INTRODUCTION

Dye-sensitized solar cells (DSSCs) have attracted considerable attention due to their environmental friendliness and low cost of production. A DSSC is composed of a nanocrystalline porous semiconductor electrode-absorbed dye, a counter electrode, and a hole transporting material. In DSSCs, the dye as a sensitizer plays a key role in absorbing sunlight and transforming solar energy into electric energy. Numerous metal complexes and organic dyes have been synthesized and utilized as sensitizers. The DSSCs sensitized by Ru-containing compounds are reaching conversion efficiencies of 11-12%, the highest efficiency reported by Chiba and Islam [1]. However, ruthenium dyes are not suitable for environmentally friendly photovoltaic systems. Ruthenium is expensive and environmentally hazardous. Ruthenium compounds are treated as highly toxic and carcinogenic. When ruthenium compounds are heated in the presence of air, they form ruthenium tetroxide, which is a highly volatile and toxic compound that damages the eyes and upper respiratory system [2]. On the other hand, natural dyes, such as pigments used in food colouring, are easily and safely extracted from plants [3]. It means that they do not require complex synthesis or toxicity test [4] and can be used in DSSCs. Since natural

dyes have low cost of synthesis and are environmentally friendly, they are considered as a viable option for dye-sensitized solar cells in future research [5].

Roy et al. indicated that when using Rose Bengal dye as sensitizer, the J_{sc} and V_{oc} of their DSSC reached 3.22mA.cm⁻² and 0.89 V, respectively, resulting in a 2.09% conversion efficiency [6]. Furthermore, Wang et al. carried out structural modification of coumarin and used the coumarin derivation dye as sensitizer in their DSSC, which provided an efficiency of 7.6% [7]. For ideal performance and excellent efficiency, the electrolyte should have high ionic conductivity so that it can transfer oxidized/reduced species to respective electrodes efficiently and should prevent back electrode reactions completely. Polyethylene oxide (PEO) has some exceptional properties of good mechanical strength, film forming properties, and excellent ability to form complexes with the ionic salts. Polymeric electrolyte is an ideal choice used in lithium ion batteries, super capacitors, photoelectrochromic display devices, and solar cells [8].

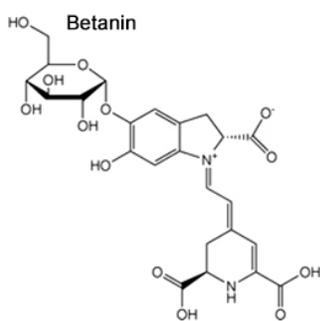
Organic liquid electrolytes dye-sensitized solar cell (DSSC) has attractive features of high energy conversion efficiency and low production cost [9, 10]. However, in the presence of traditional organic liquid electrolytes in such cells have some problems, such as lower long-term stability and a need for airtight sealing. One of the major

problems of such DSSC is the electrolyte loss caused by the leakage and volatility of the electrolyte solution that lowers the durability of the cell. Solid state dye-sensitized solar cells [DSSC] are promising due to their large potential to convert solar energy to electrical energy at low cost and their capability to solve the leakage or sealing problems that exist in liquid electrolyte dye-sensitized solar cells [11].

Natural dyes can replace synthetic dyes since they can be easily extracted from fruits, vegetables, and flowers with simple and direct chemical procedures, whereas the earlier fabrication process normally requires many steps, procedures, organic solvents, and purification procedures. In the present investigation, five natural dyes were extracted from black plum, grapes skin, beetroot, mangosteen, and raspberry. Beetroot (*Beta vulgaris*) is the main source of natural red dye, known as "beetroot red". Betanin is the main colouring compound present in red beetroot juice. Betanins are aromatic indole derivatives and have the requisite functional groups (-COOH) to bind better to the TiO₂ nanostructure [12]. Grape skin extract is

a purplish-red liquid with a fruity odor. Anthocyanins are mainly responsible for the red colour of grapes due to the highly-coloured flavylum cation [13]. Black Plum is a purple-coloured, oval-shaped tropical berry with a unique taste, flavor, and colour. *Syzygium* species are reported to be very rich in tannins, flavonoids, essential oils, anthocyanins, and others phenolic constituents. Mangosteen (*Garcinia Mangostana* L) contains a class of naturally occurring polyphenolic compounds known as xanthone. Mangostin is a natural organic compound isolated from the mangosteen plant. It is a yellow colour, crystalline solid with a xanthone core structure. Raspberries (*Rubus idaeus*) are a diverse group of flowering plants that are closely related to blackberries. The anthocyanin molecule in raspberry consists of cyanidin and pelargonidin with glucose attached at the 3-position [14]. The pictures of the black plum, grapes skin, beetroot, mangosteen, and raspberry and the molecular structures of peonidin, cyanidin, betanin, mangostin, and pelargonidin are described in Figure 1.

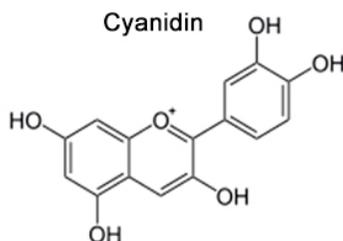
Beetroot (*Beta vulgaris*)



Black plum (*Syzygium cumini*)



Grapes skin (*Vitis vinifera*)



Mangosteen (*Garcinia Mangostana*)



Raspberry (*Rubus idaeus*)

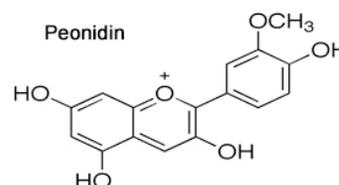
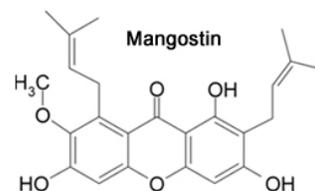
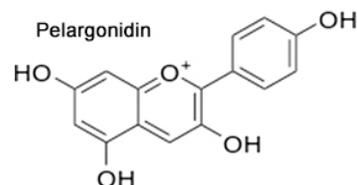


Fig 1. The pictures of the black plum, grapes skin, beetroot, mangosteen, and raspberry and the molecular structures of peonidin, cyanidin, betanin, mangostin, and pelargonidin.

These extracted dyes were characterized by FT-IR and UV-Visible absorption spectra. The synthesis and characterization of *N,N,N*-tris-(2-methoxy-naphthalene-1-yl)-*N,N,N*-triphenylbenzene-1,3,5-triamine [MNTT] as hole transporting material was investigated in our previous work. And the fabrication of DSSC using MNTT as hole transporting compound and red sandal dye as photosensitizer was also reported [15]. Here, we reported on the fabrication of solid state dye-sensitized solar cells based on MNTT as hole transporting material and different natural dyes as photosensitizers. The conversion efficiencies (η) of the solar cells were calculated by the value of photocurrent density (I_{sc}), open-current voltage (V_{oc}), and fill factor (FF).

EXPERIMENTAL

Measurements

Ultraviolet-visible (UV-Vis) spectra were recorded as diluted solution in spectroscopic grade ethanol on a UV-Vis Shimadzu 1700 using 1.0 cm length quartz tube. Infrared (IR) spectra were recorded on a Shimadzu FT-IR 8400 spectrometer as potassium bromide (KBr) disc. Fluorescence spectra were recorded as diluted solution in spectroscopic grade ethanol on a PerkinElmer LS 45 fluorescence spectrometer using 1.0 cm length quartz tube. The metal contact electrode was deposited by thermal evaporation in UHV (5×10^{-6} mbar) using a Pfeiffer evaporator (Pfeiffer PLS 500s, Labor system). Evaporation rate was 0.28 nm/s. TiO_2 was coated on a transparent conducting oxide (TCO) plate using a spin coating machine. Coating was done with a programmable spin coater SCU 2008C (Apex Instruments Co., India). The substrate was spun up to a speed of 1000 rpm for 30 s with an acceleration of 200 rpm/s. The current-voltage

(*I-V*) characteristics was monitored and measured by using a Keithley 276 source measurement unit. *I-V* measurements were generated using a self-written LabVIEW program. A solar simulator equipped with xenon lamp (AM 1.5, Solar Light Company, Model 16S-300, USA) was used as light source and light intensity was measured with a pyranometer (PMA2144, USA). The ultraviolet and infrared portion of the spectrum is eliminated by using a filter. The incident light intensity is tuned using neutral density filters (Eastman Kodak Company, USA).

Preparation of TiO_2 electrode (photoanode)

Fluorine-doped transparent conducting oxide (TCO), Deggusa P25, and titanium sulphate were purchased from Sigma Aldrich. Solvent and chemicals were used as received. Highly fluorine-doped transparent conducting oxide (TCO) films deposited on glass ($SnO_2:F$) with resistance ($30 \mu\Omega$) and thickness (1 mm) were first cleaned in a detergent solution using an ultrasonic bath for 15 minutes, rinsed with water and ethanol, and then dried. The structuring of the TCO was done using a chemical etching method. Zinc granulates were spread on the glass and reacted with HCl. The structured glass was then cleaned by ultrasonication in various solvents. Compact layers of TiO_2 were deposited on the TCO plate by spin coating method with area of 2 cm². It was prepared by precipitation of titanium sulphate using ammonium hydroxide. Then, water was added to this precipitate and stirred. 0.1 M HNO_3 was added to maintain pH (0.2) of the solution. The TCO was placed in a chamber for 1 minute and accelerated to a speed of 1000 rpm. 150 μ l of TiO_2 solution was applied onto the substrate. The solution was allowed to set for about a minute on top for good penetration into the pores. The substrate was spun up to a speed of 1000 rpm for 30 s with an acceleration of 200 rpm/s. The samples were dried for 30 minutes. After deposition, the prepared TCO/ TiO_2 was annealed at 100°C for 1 hour in air with a hot plate to achieve complete pyrolysis of organic species. The TiO_2 paste was made by mixing TiO_2 powder [Deggusa P25] with ethanol and concentrated HCl. The paste was deposited onto TCO using doctor blade technique with area of 2.0 cm² and then sintered at 350°C for 30 minutes to burn out the organic additives and to get mechanically rugged samples.

Preparation of natural photosensitizers

Extraction of dye from beetroot

The beetroot was washed with water and vacuum dried at 60°C. The cleaned vegetable was chopped and soaked in 200 mL of absolute ethanol at room temperature in the dark for one week. Then, the residual parts were removed by filtration and the filtrate was washed with hexane several times to remove any oil or chlorophyll present in the extract. Then, the filtrates were concentrated at 40°C for use as sensitizers [16].

Extraction of dye from grapes

Freshly collected grapes were soaked in 150mL of 70% ethanol and stored overnight at 4°C. The extract mixed thoroughly and filtered to remove any solid residues. Subsequently, the extracts were centrifuged for five minutes to separate all residues. The supernatant of the ethanolic extracts was gently mixed with equal volumes of petroleum ether to separate polar and nonpolar pigments [17].

Extraction of dye from black plum

The clean fresh plum fruits were dried at 40°C and soaked into a 95% ethanol solution and kept in ambient temperature. Then, solid residues were filtrated and the natural dye solutions were concentrated at 40°C and purified by chromatogram method [18].

Extraction of dye from mangosteen

The method employed for macerating the crude mangosteen rinds powder was in 80% ethanol in water at room temperature for 24 hours, with occasional stirring. The solvent was removed to yield a concentrated extract. The ethanol extract was then dissolved in 50% methanol in water and partitioned 3-4 times in a separating funnel using n-hexane for 15 minutes each to remove non-polar compounds. The n-hexane phase was removed from the funnel and ethyl acetate was added to the methanol-water phase and shaken [19]. The addition of ethyl acetate was repeated 3-4 times. The ethyl acetate phase was collected and stored in an airtight plastic container protected from light.

Extraction of dye from raspberry

The fresh raspberries were washed with water and vacuum dried at 60°C. Then, they were chopped and immersed in absolute ethanol at room temperature in the dark for one week. Then, the solids were filtered out and the filtrates were concentrated at 40°C for use as sensitizers. The dye solutions were stored in the dark and refrigerated at 4°C.

After cooling to 80°C, the TiO₂ was immersed in ethanol solution of natural dyes for 12 hours.

Preparation of hole transporting material

N,N,N-tris-(2-methoxy-naphthalene-1-yl)-*N,N,N*-triphenylbenzene-1,3,5-triamine [MNTT] was synthesized and characterized [15]. The structure of MNTT as described in Figure 2.

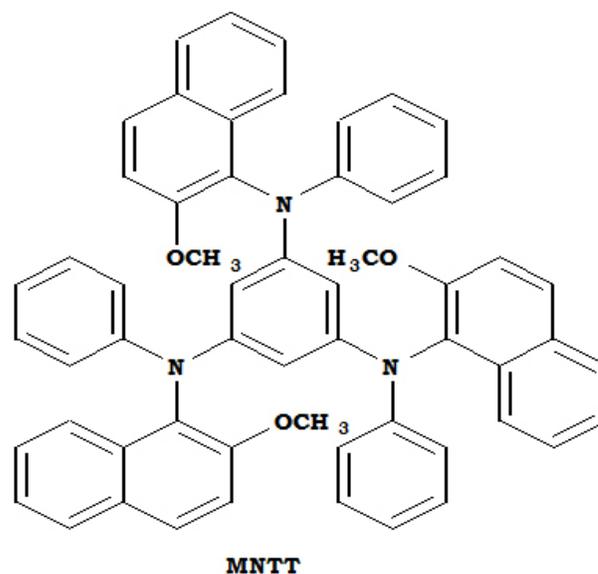


Fig 2. Structure of hole transporting material [MNTT]

In these DSSC, HTMs in THF (~800 nm) were deposited on the cell by spin coating method. The organic cells were kept overnight to allow maximum penetration of HTM in TiO₂.

Silver counter electrode

The metal electrode silver (200nm) was coated on HTM of DSSC by thermal evaporation.

RESULTS & DISCUSSION

Characterization of dyes

IR spectra of dyes

Black plum dye: 3415 cm⁻¹ (O-H stretching); 2933 cm⁻¹ (Aromatic C-H stretching); 2850 cm⁻¹ (methyl C-H stretching); 1448 cm⁻¹ (methyl C-H bending); 1259 cm⁻¹ (C-O-C stretching); and 1058 cm⁻¹ (C-O stretching).

Grapes dye: 3386 cm^{-1} (O-H stretching); 2937 cm^{-1} (aromatic C-H stretching); 1460 cm^{-1} (methyl C-H bending); and 1060 cm^{-1} (C-O stretching).

Raspberry dye: 3359 cm^{-1} (O-H stretching); 2960 cm^{-1} (aromatic C-H stretching); 1461 cm^{-1} (methyl C-H bending); and 1072 cm^{-1} (C-O stretching).

Mangosteen dye: 3404 cm^{-1} (O-H stretching); 2933 cm^{-1} (aromatic C-H stretching); 2852 cm^{-1} (methyl C-H stretching); 1448 cm^{-1} (methyl C-H bending); 1261 cm^{-1} (C-O-C stretching); and 1060 cm^{-1} (C-O stretching).

Beetroot dye: 3421 cm^{-1} (O-H stretching); 2925 cm^{-1} (aromatic C-H stretching); 1448 cm^{-1} (methyl C-H bending); 1053 cm^{-1} (C-O stretching); and 1263 cm^{-1} (C-N stretching).

UV- Vis spectra of dyes

We attempted to use five kinds of colourful natural dyes as sensitizers for DSSCs. Table 1 lists the UV-vis absorption data of the dyes extracted with ethanol. To investigate the absorption process of sunlight into dye, the UV-Vis measurement was performed between wavelength of 200nm and 700nm. The maximum absorption of each dye is listed in Table 1. These five dyes have better absorption features in the UV light zone. The absorption peaks of beetroot dye are at 478nm and 540nm. The anthocyanin dye extracted from different natural photosensitizer has different absorption peaks. The anthocyanin dye from grapes shows absorption at 529nm, anthocyanin from raspberry shows two absorption peaks at 207 nm and 279 nm, and the absorption peaks of anthocyanin extracted from black plum are at 553 nm and 662 nm. The dye extracted from mangosteen absorbs at 378 nm. The ethanol extract of mangosteen pericarp showed various colours, whereas no obvious maximum absorption peak in the visible light region was observed. This result can be attributed to the superposition of absorption peaks. All dyes contained natural phenolic compounds. The chemical adsorption of these dyes is

generally accepted because of the condensation of alcoholic-bound protons with the hydroxyl groups on the surface of nanostructured TiO_2 [17].

Fluorescence spectra of dyes

Among the five natural dyes, the dyes extracted from raspberry and mangosteen show fluorescence peaks. Raspberry has three fluorescence peaks and fluorescence maximum at 636 nm. Mangosteen has two fluorescence peaks and fluorescence maximum at 763 nm. The light harvesting effect of dye plays an important role in capturing the photons and generating the electron/hole pair, as well as transferring them to the interface of the semiconductor and the electrolyte, respectively. The fluorescence material was absorbed on the TiO_2 photo-electrode with sensitizers in dye-sensitized solar cell (DSSC) to enhance the photon-to-current efficiency. The improved light harvesting efficiency, which was achieved by the judicious choice/design of fluorescence material and sensitizing dyes, enhances the photovoltaic performance of the DSSCs.

Photoelectrochemical properties of DSSC sensitized with natural dyes

The dye sensitized solar cells based on raspberry, mangosteen, beetroot, black plum, and grapes are DSSC 1, DSSC 2, DSSC 3, DSSC 4, and DSSC 5, respectively. Photovoltaic tests of DSSC using these natural dyes as sensitizers were performed by measuring the current-voltage (I-V) characteristics under irradiation with white light (100 mWcm^{-2} from 300 W solar simulator). The performance of natural dyes as sensitizers in DSSC was evaluated by short circuit current (I_{sc}), open circuit voltage (V_{oc}), fill factor (FF), and energy conversion efficiency (η). The photoelectrochemical parameters of the DSSC are listed in Table 1.

Table 1. Photo-electrochemical properties of dye sensitizes solar cell

Sample	Absorption maximum [nm]	ISC [mA/cm^2]	VMAX [V]	IAX [mA/cm^2]	FF [%]	η [%]
DSSC1	534	1.88	1.5	1.25	0.50	0.94
DSSC2	378	0.49	1.75	0.35	0.63	0.31
DSSC3	478, 540	0.35	1.75	0.23	0.58	0.20
DSSC4	553, 662	0.27	1.75	0.20	0.65	0.18
DSSC5	529	0.09	1.75	0.06	0.58	0.05

CONCLUSION

Different dyes show different photo conversion efficiency; that is, 0.94, 0.31, 0.20, 0.18, and 0.05 are DSSC 1, DSSC 2, DSSC 3, DSSC 4, and DSSC 5, respectively. Maximum 0.94% efficiency is obtained for raspberry dye.

Anthocyanins extracted from black plum, grape skin, and raspberry consist of an aromatic ring bonded to a heterocyclic ring containing an oxygen atom which in turn is linked by a C–C bond to a third aromatic ring; so, it may also be described as a C6–C3–C6 skeleton. The extended p conjugation, as well as the presence of positive charge and free –OH groups, allows the anthocyanins to absorb light in the visible region leading to a large variety of dye colours. The difference between anthocyanins of different types is mainly due to the number of hydroxyl and/or methoxy groups in the molecule. Due to this chemical complexity, there is a large variety of anthocyanins in nature and three anthocyanidins, i.e., cyanidin, pelargonidin, and peonidin are presented in our dyes. The number of hydroxyl and methoxyl groups determines the intensity, type, and stability of anthocyanins' colour. Colour stability refers to the capacity of the molecule (dye) to maintain its colour properties. Generally, predominance of hydroxyl groups on the aromatic skeleton of anthocyanins gives rise to an intense blue colour while a red colour is observed when methoxyl groups prevail instead. Anthocyanine strongly absorb on the TiO₂ as a result of a highly stable Ti4+–anthocyanin complex. The maximum absorption shown is grape dye, but raspberry dye (pelargonidin) has high efficiency. This is because raspberry dye shows fluorescence.

Betanins possess the carboxyl functional groups (–COOH) that is a key requisite to bind to the TiO₂ nanostructure via the formation of ester-type linkage. Upon absorption of the dye onto the TiO₂ surface, the proton of the carboxyl group is transferred to the oxide (e.g., TiO₂). Contrarily, for anthocyanins the absorption onto TiO₂ requires the presence of two ortho-hydroxyl-groups that, on one hand leads to strong electronic coupling and a rapid electron transfer from the dye to the TiO₂ and on the other hand also assists the back electron transfer (i.e., recombination process).

Mangostin extracted from mangosteen pericarp shows lesser absorption, but it contains methoxy and hydroxyl group that can help to better absorption on TiO₂. Mangosteen has two fluorescence peaks and fluorescence maximum at 763 nm. The fluorescence material was absorbed on the TiO₂ photo-electrode to enhance the photon-to-current efficiency.

Vegetable dyes are available in large quantity, easily and safely extracted from fruits, low in cost, and non-toxic. All these factors make the use of these natural products an intriguing challenge in the development of cheap and commercially available DSSCs. Organic dye-sensitized solid state solar cells were fabricated using synthesized HTM and different natural dyes. Different dyes show different photo conversion efficiency that varies from 0.94 to 0.05%. From the results, we can say that DSSC fabricated using raspberry shows high efficiency. In our previous report, the efficiency DSSC fabricated using red sandal dye was 0.25%. Raspberry, mangosteen, and red sandal show fluorescence peaks. Raspberry has three fluorescence peaks and fluorescence maximum at 636 nm. Mangosteen has two fluorescence peaks and fluorescence maximum at 763 nm. Red sandal has one fluorescence peak and fluorescence maximum at 536 nm. The fluorescence material was absorbed on the TiO₂ photo-electrode with sensitizers in dye-sensitized solar cell (DSSC) to enhance the photon-to-current efficiency.

Instead of the ruthenium dye and back electrode gold, we have used the natural dyes and silver. These are the two major differences we adopted. We would like to highlight the total cost effect of the cell, which will be down to nearly 40% of a similar type of fabricated cell. The conversion of the non-conventional energy, even if for a very low percentage, is an advancement for mankind. The use of a lesser amount of chemicals is an added advantage when the natural dye is applied. This will be an introductory step to the green synthesis. The simple extraction procedure, low cost, and environmentally friendly natural dyes are promising sources of sensitizers for DSSCs.

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