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Dye-Sensitized Solar Cells Using *Carissa carandas* Linn. Fruits Extract as Dye Sensitizer†

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Dye-sensitized solar cells (DSSCs) are a promising light harvesting material. The dye sensitizers using as solar energy absorber play a crucial on the conversion efficiency. There are two types of dye sensitizers; organic dyes and inorganic dyes. The organic dyes, derived from nature, have some advantages over the inorganic metal complex dyes as follow; (1) higher absorption coefficients; (2) wide range of absorption spectrum; (3) low-cost and environmentally friendly. In this study, the natural dyes extract from *Carissa carandas* Linn. fruits was used as sensitizers to fabricate dye-sensitized solar cells. The UV-visible adsorption characteristics of natural dye solution were shown to absorb in visible region at λ_{max} 537.92 nm. The light conversion efficiency (η) of dye-sensitized solar cells reached 1.19 % with a short-circuit current density (I_{SC}) of 3.08 mA cm^{-2}, an open circuit voltage (V_{OC}) of 0.64 V and a fill factor (ff) of 0.56.

Keywords: Dye-sensitized solar cells, Photo sensitizers, Light harvesting material.

INTRODUCTION

Solar energy is one of the most interesting energy sources because it is clean and environmental friendly. In the last decade, the new molecular photovoltaic (PV) materials, called dye-sensitized solar cells (DSSCs), have emerged and could be alternative low-cost solar cells in the near future. They are designed for light harvesting which enable of using a dye sensitizer (DS) on semiconductor. Dye-sensitized solar cells are composed mainly of non-toxic materials and inexpensive technology for high efficient solar cells[1,2]. Dye-sensitized solar cells had gained interest in the first time after Grätzel[3] and Nazeeruddin *et al.*[4] reported a high solar energy for electricity conversion which has efficiency up to 11 % with ruthenium bipyridyl dyes. However, the dye-sensitized solar cells based on ruthenium complexes are quite expensive for cost-conversion efficiency because the noble metal ruthenium is a limited resource and the synthesis processes of complexes are very complicated and difficult to achieve their purities. Furthermore, ruthenium complexes containing heavy metals are not environmentally friendly[5].

Natural dyes can be easily extracted by simple procedure from the leaf, flower and fruit of plants which are displayed in various colours and harvested light in the visible region[6,7]. Recently, several groups have been working on natural dye sensitizers to overcome the prohibitive issues of ruthenium complexes. The advantages of natural dyes as photo sensitizers are large absorption coefficients, high light harvesting efficiency, no resource limitations, low cost, simple prepared and low environmental impacts[8,9]. Several natural dyes including flavonoids, cyanins, tannins, carotenes, anthocyanins and chlorophylls have been reported as sensitizers in dye-sensitized solar cells in recent years[10-12]. These results indicated that natural dyes would be a promising type of sensitizers for dye-sensitized solar cells.

In this paper, natural dyes extract from *Carissa carandas* Linn. fruits was used as sensitizers in dye-sensitized solar cells. The natural dyes were purified using solid phase extraction (SPE) and characterized by UV-visible absorption spectra. The photoelectrochemical properties of the dye-sensitized solar cells using these extracts as sensitizers were investigated depending on open circuit voltage (V_{OC}), short-circuit current density (I_{SC}), maximum power point (MPP), fill factor (ff) and overall energy conversion efficiency (η).

EXPERIMENTAL

All chemicals and transference conductive glass plates (Fluorine-doped tin oxide; FTO, sheet resistance 8 Ω per cm^2) were purchased from Sigma-Aldrich Company. UV-visible spectra were recorded on UV-visible Spectrometer Perkin-

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Elmer Lambda 20. Stimulate sunlight produce from xenon arc lamp (Osram Sylvania Inc.). Current-Voltage (I-V) characteristics of solar cells were investigated by Sourcemeter (Keithley Instruments Inc.).

Preparation of dye sensitizers: Di-tetrabutylammonium *cis-bis*(isothiocyanato)*bis*(2,22-bipyridyl-4,42-dicarboxylato) ruthenium(II) (N-719) was prepared in a 0.3 mM solution in absolute ethanol and used as a standard dye sensitizer in all measurements. The ethanol extract of natural dye was prepared by soaking fresh fruits of *C. carandas* Linn. 100 g in 500 mL of ethanol and maintained for 24 h at room temperature. The crude extract was passed through solid phase extraction (SPE) cartridges containing octadecylsilane bonded phase. After cleanup, the eluent was evaporated *in vaccuo* to a give dark red-purple solid. The natural dye solution was prepared by dissolving 1 g of natural dye extract in 100 mL of absolute ethanol[13].

Preparation of thin-film TiO_2 working electrodes: Titanium dioxide nanoparticle powder (6 g), ethyl cellulose (2 g), polyethylene glycol (2 g) (F.W. 20,000) and Triton X-100 were added into a mortar and ground to form slurry[14]. Porous TiO_2 films (1 cm^2) were prepared by a screen printing technique by spreading onto fluorine-doped SnO_2 (FTO) conducting glass which was pre-cleaned ultrasonically in ethanol, then rinsed with acetone and dry at 120 °C. This procedure was repeated for 5 times, each coating was allowed to dry at room temperature. The film was allowed to dry overnight in a vacuum desiccator. The dried film was then sintered at 450 °C for 30 min at a heating rate of 5 °C min^{-1}, cooled down to 80 °C[15] and then kept in a desiccator.

Preparation of platinum counter electrode: Platinum counter electrode (1 cm^2) was prepared by deposition of a paste containing 5 mM H_2PtCl_6 in α-terpineol using a screen printing technique for 5 times on a previously cleaned fluorine-doped SnO_2 conducting glass[16]. After air-drying at room temperature, the plates were sintered in a furnace at 400 °C for 0.5 h at a heating rate of 20 °C min^{-1} and the warm plates were moved into a desiccator.

Fabrication and measurements efficiency of dye-sensitized solar cells (DSSC): The TiO_2 electrodes were immersed into the dye-sensitized solution and kept in a dark room overnight at room temperature allowing the adsorption of nanocrystalline TiO_2. After that, the electrodes were rinsed with ethanol and dried. Counter electrode was directly placed on top of the dye-adsorbed TiO_2 electrode. Surlyn polymer sheet was used as a spacer to separate TiO_2 surface and counter electrode to avoid direct contact. Electrolyte solution consisted of a solution of 0.1 M lithium iodide, 0.05 M iodine and 0.5 M 4-*tert*-butylpyridine in anhydrous acetonitrile as solvent was filled to the cell[17]. The current–voltage performance of the dye-sensitized solar cell was measured by exposing with simulated AM 1.5 irradiation with an incident power density of 100 mW cm^{-2}.

RESULTS AND DISCUSSION

The UV-visible spectra of ethanolic solution of natural dye and N719 ruthenium complex were measured and the absorption regions of the dye solutions are shown in Fig. 1.

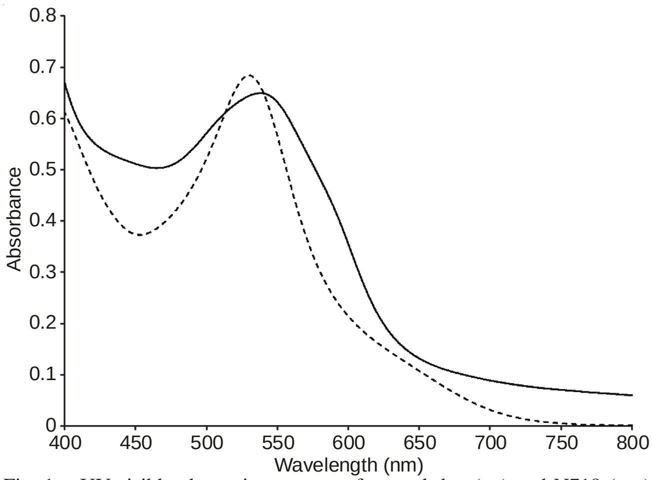

Fig. 1. UV-visible absorption spectra of natural dye (—) and N719 (----) in absolute ethanol

N719 dye solution absorbed the light in visible region at λ_{max} 530.09 nm. The long wavelength absorption of N719 dye in the form of ruthenium complex was due to a metal-to-ligand charge-transfer (MLCT) energy[18]. The natural dye was a metal-free dye, which consisting a conjugate π-bond system. The absorption for this system was due to the strong π to π* molecular transitions[19]. The absorption maximum for natural dye was observed at 537.92 nm. It can be seen that the absorption ranges of the two dyes are close to each other.

Measurements of dye-sensitized solar cells: The dye-sensitized solar cells measurements were done by exposing with simulated AM 1.5 irradiation with an incident power density of 100 mW cm^2 (P_{in}). The current–voltage of the dye-sensitized solar cells measurements under simulated irradiation tests were automatically recorded by a computer. The results were obtained as I–V curve by the plot of a short-circuit current density (I_{SC}) against an open circuit voltage (V_{OC}), giving the maximum power point (MPP). The I–V curve of dye-sensitized solar cells using N719 and natural dye are shown in Fig. 2.

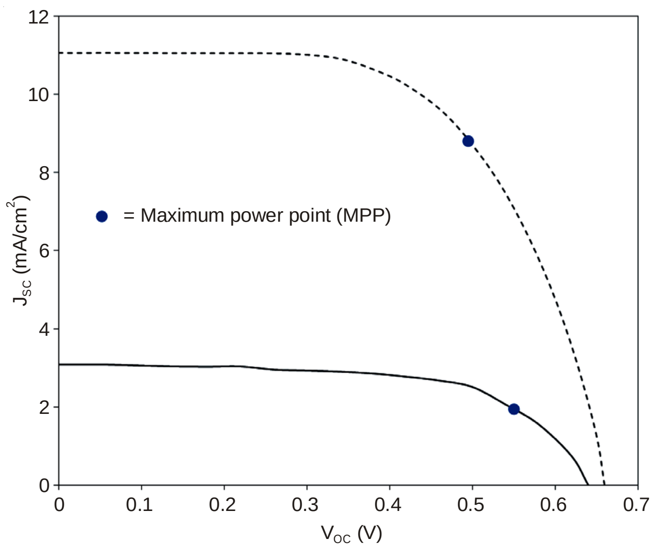

Fig. 2. I-V Curve of dye-sensitized solar cells measurement using natural dye (—) and N719 (---) as dye sensitizer

The values of fill factor (ff) and overall energy conversion efficiency (η) were calculated by the following equations[20].

$$\text{Fill factor (ff)} = \frac{V_{max} \cdot I_{max}}{V_{OC} \cdot I_{SC}} \quad (1)$$

$$\eta\,(\%) = \frac{V_{OC} \cdot I_{SC} \cdot ff}{P_{in}} \times 100 \quad (2)$$

The performance of the natural sensitized and inorganic N719 solar cells were measured. The results are shown in Table-1.

TABLE-1
PHOTOVOLTAIC PERFORMANCE DYE-SENSITIZED SOLAR CELLS UNDER SIMULATED IRRADIATION SOURCE (AM 1.5, 100 mW cm^{-2})

Dye	I_{SC} (mA cm^{-2})	V_{OC} (V)	ff	η (%)
N719	11.05	0.66	0.64	4.66 ± 0.01
Natural dye	3.08	0.64	0.56	1.10 ± 0.05

The performance of dye-sensitized solar cells of N719 using simulated irradiation source (AM 1.5, 100 mW cm^{-2}) gave a I_{SC} of 11.05 mA cm^{-2}, V_{OC} of 0.66 V and a fill factor of 0.64 with the corresponding conversion efficiency (η) of 4.66 %. Under the same condition, the conversion efficiency of sensitized solar cells using natural dye was obtained as 1.10 % with I_{SC} of 3.08 mA cm^{-2}, V_{OC} of 0.64 and a fill factor of 0.56. The reasons for the higher efficiency of N719 compared with natural dye due to a strong charge-transfer (CT) absorption in the whole visible range, long excited lifetime and highly efficient metal-to-ligand charge transfer (MLCT)[21].

Conclusion

The natural dyes extract obtained from the fruits of *Carissa carandas* Linn. was used as sensitizers in dye-sensitized solar cells. The absorption spectrum of natural dye solution strongly absorbed visible light between 450 and 600 nm and λ_{max} was observed at 537.92 nm which nearly maximum absorption spectrum of N719 ruthenium complex. The dye-sensitized solar cells were fabricated by sandwiching a polymer spacer between a dye-coated nanoporous TiO_2 electrode and a platinum counter electrode. The conversion efficiency of the sensitized cells using the natural dye was comparable with N719 ruthenium complex sensitized cells. The N719 and natural dye sensitized cells gave conversion efficiency (η) of 4.66 % and 1.10 % respectively.

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