

Fabrication of Dye-Sensitized Solar Cells Using Dried Plant Leaves

Sofyan A. Taya*[‡], Taher M. El-Agez^{1*}, Monzir S. Abdel-Latif^{**}, Hatem S. El-Ghamri*,
Amal Y. Batniji^{*}, Islam R. El-Sheikh^{*}

*Physics Department, Islamic University of Gaza, P.O.Box 108, Gaza, Palestinian Authority;

**Chemistry Department, Islamic University of Gaza, P.O.Box 108, Gaza, Palestinian Authority.

(staya@iugaza.edu.ps, telagez(@iugaza.edu.ps, mlatif(@iugaza.edu.ps, hghamri(@iugaza.edu.ps, abatniji(@iugaza.edu.ps, irshaikh(@iugaza.edu.ps)

[‡]Corresponding Author; Sofyan A. Taya , 00972 8 2644400 (ext 1025), staya@iugaza.edu.ps

Received: 22.03.2014 Accepted: 24.04.2014

Abstract- In this work, dye-sensitized solar cells (DSSCs) based on natural dyes extracted from six dried plant leaves were fabricated. The extracts were characterized by UV-Vis absorption spectroscopy. TiO₂ nanopowder film on FTO-coated glass was used as anode electrode. The photovoltaic properties of the fabricated DSSCs were investigated under an incident irradiation of 100 mW/cm². The best performance was found for the DSSC sensitized with Jasminum Grandifolium with a solar energy conversion efficiency of 0.335%. The performance of the fabricated cells was investigated at various pH values of flora dye solutions. Moreover, the photovoltaic properties were studied in different extraction solvents.

Keywords- dye sensitized solar cells, natural dyes, plant leaves, TiO₂.

1. Introduction

Energy technology is one of the most important technologies, since the demand for energy is growing day by day. Moreover, fossil fuels rapid resource depletion and environmental pollution have increasingly become a worldwide concern in the past few decades. Thus, there is an urgent need of sustainable energy resources, such as solar energy. Enhancing the efficiency of solar energy use has become an important research issue. Despite of the recent developments in solar energy technology, the silicon solar cells have not been popularized because of their high cost and the high tech of silicon-based raw material production.

In recent years, dye-sensitized solar cells (DSSCs) have been developed as the third generation of solar cells [1]. DSSCs have received increasing attention from researchers because of their environmental friendliness, flexibility, low-cost and abundant materials, as well as high power conversion efficiencies under cloudy and artificial light conditions. DSSCs are devices that convert visible light into electricity through the sensitization of wide-band-gap semiconductors. The sensitization process is performed using natural as well as synthetic dyes [1-15]. The principle of operation of DSSCs is based on exciton creation due to

photon- dye molecule interaction. The generated excitons split to form electrons and holes. Electrons are attracted toward the semiconductor and then to FTO while holes are transported in the other direction. The photoelectric conversion efficiency of DSSCs was reported to reach 10-11% [2,3]. The photoelectrode is one of the most important elements for obtaining high photoelectric conversion efficiency [4]. The sensitizing dye, as a part of the photoelectrode, plays a key role in absorbing sunlight and transforming solar energy into electrical energy.

Although many metal complexes have a number of interesting features such as good absorption and highly efficient metal-to-ligand charge transfer, high cost and complicated preparation techniques of the efficient ones gave more attention toward natural dyes, which can be easily extracted from flowers, leaves, seeds, and fruits. Due to their low cost, abundance, non-toxicity, and complete biodegradation, natural dyes have received an increasing interest.

Several natural dyes have been utilized as sensitizers for DSSCs. El-Agez et al. studied natural dyes extracted from fresh and dried plant leaves and found that spinach oleracea extract has a better performance after drying where the

efficiency of the cell prepared with TiO₂ thin film layer reached 0.29% [5,6]. Plant seeds have been used as sensitizers and it was found that DSSCs sensitized with the extracts of onion, rapa, and *Eruca sativa* seeds have efficiencies of 0.875%, 0.86%, and 0.725%, respectively [7]. In 2008, red Sicilian orange juice dye was used as a sensitizer and a conversion efficiency of 0.66% was reported [8]. Rosella was used as a sensitizer for DSSCs with efficiency of 0.70% [9]. Roy et al. reported that DSSCs sensitized with Rose Bengal dye can have conversion efficiency of 2.09% [10]. The modified structure of coumarin derivative dye was proposed by Wang et al. which provided an efficiency of 7.6% [11]. J. Etula [12] and R. Ahmadian [13] studied the structure and the concentration of anthocyanins, respectively, in several natural dyes used as sensitizers for DSSCs. It was hypothesized, that natural dyes with higher anthocyanin concentration, such as those extracted from blueberry and black raspberry, have higher fill factors and efficiency. Chlorophyll (A) structure in *Punica granatum* peel extract gave a solar cell with 1.86% conversion efficiency [14]. In general, natural pigments, such as anthocyanins, carotenoids, and chlorophylls, have several advantages over rare metal complexes for DSSC sensitization.

In this paper, the extracts of six dried plant leaves were used as sensitizers for dye-sensitized solar cells. The extracts were characterized by UV-Vis absorption spectroscopy. TiO₂ nanopowder was used as a semiconducting material. The photovoltaic properties of the DSSCs were carried out under an incident irradiation of 100 mW/cm². The effects of pH and different solvents of the flora extract on the photovoltaic properties were investigated.

2. Experiment

2.1. Preparation of natural dye sensitizers

Six plant leaves were used: *Flora*, *Jasminum Grandifolium*, *Rosa damascena*, *Carthamus tinctorius*, *Petroselinum crispum*, and *Spinacia oleracea*. These leaves were left to dry for seven days. One gram of the dried leaves fine powder was added to 10 ml of ethanol as a solvent and left for 24 h. The solutions were then filtered and concentrated at 60 °C to a final volume of 3 ml.

2.2. Assembly of the cell

FTO conductive glass sheets with resistance of 15 Ω/cm² were cut into small pieces each of dimensions 1cm×1cm. An ultrasonic bath was used to clean the samples. The thickness and area of the TiO₂ layer were restricted by fixing plastic adhesive tape on three sides of the FTO sides. TiO₂ nanopowder (10-25 nm) purchased from US Research Nanomaterial, Inc, USA, was used as a semiconducting material. Equal weights of TiO₂ nanopowder and polyethylene glycol were mixed and grinded in a mortar for half an hour to form a homogeneous paste. Doctor-blade technique was used to spread TiO₂ thin films of area of 0.25 cm² onto the FTO sheets. The TiO₂ films were left to dry at 60 °C for 30 min. Finally, the TiO₂ films were sintered at

450 °C for 30 min [15], cooled down to a temperature of 60 °C, and then immersed in the solutions of the dried plant leaves extracts for one day for adequate dye anchoring on the TiO₂ porous films. TiO₂ film and a conductive glass sheet plated with platinum (Pt) were used as anode and cathode electrodes, respectively. The DSSC was assembled by filling a liquid electrolyte between the electrodes. The electrolyte solution was composed of 2 ml acetonitrile (ACN), 8 ml propylene carbonate (p-carbonate), 0.668 gm (LiI), and 0.0634 gm (I₂). Then the two electrodes were clipped together to form the DSSC.

2.3. Measurements

UV-Vis spectrophotometer (GENESYS 6, Thermo Scientific, USA) was used to measure the absorption spectra of all dye extracts in the spectral range from 400 nm to 800 nm. National Instruments data acquisition card (USB NI 6251) in combination with Lab VIEW program was used to carry out the current-voltage characteristic curves of the fabricated DSSCs. The current-voltage measurements were established at 100 mW/cm² irradiation using a high-pressure mercury arc lamp.

3. Results and Discussions

3.1. Absorption of Natural Dyes

The UV-Vis absorption spectra of the extracts of *Flora*, *Jasminum Grandifolium*, and *Rosa damascena* are shown in Fig. 1. As can be seen from the figure there is an absorption peak at about 665.80nm for the extract of *Flora*. The absorption spectra of *Jasminum Grandifolium* and *Rosa damascena* show peaks at 658.1 nm and 420.51 nm, respectively.

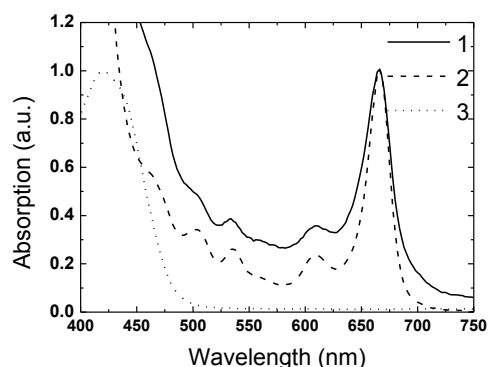


Fig. 1. Absorption spectra of the extracts of *Flora* (1), *Jasminum Grandifolium* (2), and *Rosa damascena* (3).

3.2. Photoelectrochemical Properties

The current density (J)-voltage (V) characteristic curves of the fabricated DSSCs were carried out under illumination with white light of intensity 100 mW/cm². Figure 2 illustrates the J-V characteristic curves for the DSSCs sensitized with *Flora* (1), *Jasminum Grandifolium* (2), *Rosa damascena* (3), *Carthamus tinctorius*(4), *Petroselinum crispum*(5), and *Spinacia oleracea*(6). The most significant

parameters for determining the DSSC performance are the short circuit current density, J_{sc} , and open circuit voltage, V_{oc} . J_{sc} and V_{oc} were determined from the J-V curve interceptions with y- and x-axes, respectively. The DSSCs output power was calculated as $P = JV$ using the J-V data corresponding to each cell. From the output power data, the maximum power point was determined for each cell. The current density and voltage of maximum power point were denoted as J_m and V_m . The fill factor (FF) and the cell efficiency (η) were then calculated using these parameters [15]. All results were summarized in the Table 1. It is clear that the short-circuit current ranged from 0.269 to 1.278 mA/cm² for the DSSCs sensitized with Carthamus tinctorius

and Flora, respectively. The short-circuit currents obtained for the DSSCs sensitized with Jasminum Grandifolium, Rosa damascena, Spinacia Oleracea, Petroselinum crispum were, respectively, 1.198, 0.276, 0.66 and 0.33 mA/cm². The open-circuit voltage ranged from 0.610 V for the DSSC sensitized with Petroselinum crispum to 0.665 V for that sensitized with Jasminum Grandifolium. The conversion efficiency of the fabricated cells ranged from 0.057% for the DSSC sensitized with Rosa damascena to 0.335% for that sensitized with Jasminum Grandifolium. Therefore, it is clear that Jasminum Grandifolium is a strong candidate as a sensitizer for efficient DSSCs.

Table 1. Photocurrents (J_{sc} , J_m), photovoltages (V_{oc} , V_m), fill factor, and overall energy conversion efficiency for the fabricated DSSCs.

| Dye | J_{sc} (mA/cm) | V_{oc} (V) | J_m (mA/cm ²) | V_m (V) | FF | η % |
|-----------------------|---------------------|-----------------|--------------------------------|--------------|-------|-------------|
| Flora | 1.278 | 0.651 | 0.85 | 0.444 | 0.377 | 0.314 |
| Jasminum Grandifolium | 1.198 | 0.665 | 0.87 | 0.462 | 0.401 | 0.335 |
| Rosa damascena | 0.276 | 0.617 | 0.165 | 0.42 | 0.069 | 0.057 |
| Carthamus tinctorius | 0.269 | 0.619 | 0.161 | 0.436 | 0.070 | 0.058 |
| Petroselinum crispum | 0.33 | 0.610 | 0.18 | 0.424 | 0.076 | 0.063 |
| Spinacia oleracea | 0.667 | 0.625 | 0.46 | 0.466 | 0.214 | 0.178 |

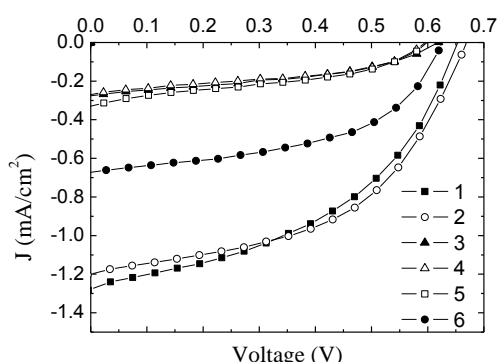


Fig. 2. J-V characteristic curves for the DSSCs sensitized with Flora (1), Jasminum Grandifolium (2), Rosa damascena (3), Carthamus tinctorius(4), Petroselinum crispum(5) and Spinacia oleracea(6), under the illumination with white light of intensity 100 mW/cm²

The effect of pH of flora dye solution on the absorption spectrum of the flora extract was investigated. Figure 3 shows the optical absorption spectra of the various flora dye solutions, whose pH was controlled by adding hydrochloric acid or sodium hydroxide. Seven different pH values ranging from 2.14 to 12.22 were examined. When pH was 5.58, a relatively high absorption peak was observed at about 655.5 nm. On the other hand, high absorption peak was observed at around 450 nm when pH was 12.22, which suggests the acid-base nature of the extract. The J-V characteristic curves of the fabricated DSSCs using various flora dye solutions with

different pH values are shown in Fig. 4. The short circuit currents, open circuit voltages, fill factors and efficiencies extracted from these curves are summarized in Table 2. As presented in this Table, the conversion efficiency of the DSSCs fabricated using flora dye were highly dependent on pH, where an increase in the efficiency of about 100% was achieved by controlling the pH. The reason of the high value of the conversion efficiency of the DSSC sensitized with flora dye with a pH of 5.58 may be attributed to the increase of the optical absorption of the dye, as shown in Fig. 3. The small shifting of the wavelength of the optical absorption peak (see Fig. 1) is considered to be due to the change of the molecular structure of the dye as a result of change in pH. This is in agreement with the previous report of formation of flavylium ion form is favorable [16] at low pH.

Figure 5 shows J_{sc} , V_{oc} , FF, and η of the fabricated cells as a function of the pH of the flora dye solution. It is obvious that there is a significant impact of the pH on the cell performance. There is an optimum value of the pH (about 5.6) at which all the photoelectrochemical parameters of the cell were improved.

Table 2. Characteristics of the DSSCs fabricated using various flora dye solutions whose pH was controlled by adding hydrochloric acid or sodium hydroxide

| pH | J_{sc} (mA/cm) | V_{oc} (V) | FF | η % |
|------|---------------------|-----------------|-------|-------------|
| 2.14 | 0.855 | 0.610 | 0.322 | 0.167 |
| 2.97 | 0.916 | 0.605 | 0.324 | 0.179 |
| 4.41 | 0.980 | 0.599 | 0.357 | 0.209 |
| 5.58 | 1.278 | 0.651 | 0.377 | 0.314 |

| | | | | |
|-------|-------|-------|-------|-------|
| 6.64 | 0.754 | 0.630 | 0.364 | 0.172 |
| 7.20 | 0.733 | 0.635 | 0.362 | 0.167 |
| 12.22 | 0.692 | 0.646 | 0.369 | 0.164 |

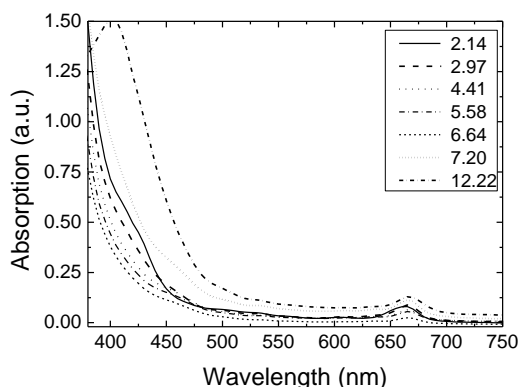


Fig. 3. Absorption spectra of various flora dye solutions whose pH was controlled by adding hydrochloric acid or sodium hydroxide.

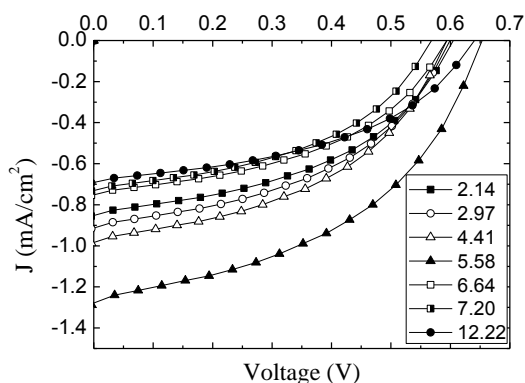


Fig. 4. J-V characteristic curves for the DSSCs sensitized with flora dye whose pH was controlled by adding hydrochloric acid or sodium hydroxide.

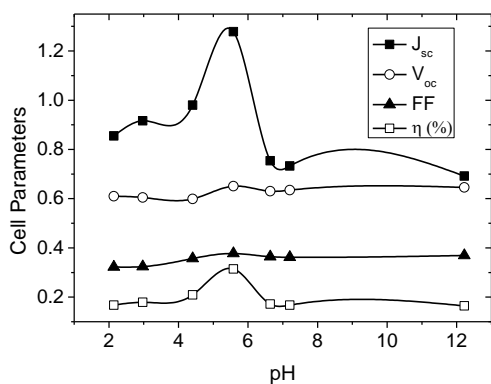


Fig. 5. DSSC parameters versus the pH of the dying solution.

Finally, the effect of Flora dye extraction solvent on the performance of the fabricated DSSCs is investigated. The

photovoltaic properties of the DSSCs sensitized with the dyes extracted from flora with various solvents were studied by measuring J–V curves as shown in Fig. 6. The corresponding photoelectrochemical parameters are listed in Table 3. As observed, the efficiencies of the DSSCs fabricated using Acetone, Ethylene glycol, Polyethylene glycol, Water, and Ethyl alcohol solvents are 0.207% and 0.150%, 0.03%, 0.05%, and 0.314%, respectively. Moreover, Table 3 shows that the V_{oc} , J_{sc} , and η of the extracts using ethyl alcohol and acetone solvents are higher than those of the extracts with polyethylene glycol and water. The reason for this behavior is unclear and requires further investigation.

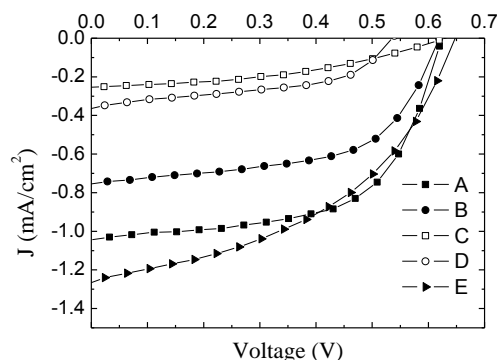


Fig. 6. J-V characteristic curves for the DSSCs sensitized with flora dye extracted using Acetone (A), Ethylene glycol (B), Polyethylene glycol (C), Water (D), and Ethyl alcohol (E) solvents.

Table 3. Effect of extracting solvent on DSSC efficiency using various flora dye with four different solvents.

| Solvent | J_{sc} (mA/cm ²) | V_{oc} (V) | FF | η % |
|---------------------|--------------------------------|--------------|-------|----------|
| Acetone | 1.04 | 0.619 | 0.322 | 0.207 |
| Ethylene glycol | 0.75 | 0.619 | 0.324 | 0.150 |
| Polyethylene glycol | 0.25 | 0.519 | 0.197 | 0.030 |
| Water | 0.37 | 0.539 | 0.254 | 0.050 |
| Ethyl alcohol | 1.27 | 0.651 | 0.377 | 0.314 |

4. Conclusion

Dye sensitized solar cells based on natural dyes extracted from dried plant leaves were fabricated using TiO₂ semiconducting material. The extracted dyes were characterized by UV-Vis absorption spectra. The J-V characteristic curves were measured and the photoelectrochemical properties were investigated. The highest conversion efficiency was obtained for the DSSC sensitized with the extract of *Jasminum Grandifolium*. The pH of the dye solution was found to have a significant effect on the performance of the cell. There is an optimum pH at which the efficiency of the cell peaks. Moreover, it was found that the extraction solvent has a crucial impact on the

fabricated cell parameters. When using the extract of flora, ethyl alcohol solvent was found to have the highest efficiency.

Acknowledgements

The authors would like to express gratitude to the scientific research affairs at the Islamic university of Gaza for the financial support of this work under the research grant for the academic year 2012-2013.

References

- [1] B.O'Regan, M. Grätzel, "A low-cost, high-efficiency solar cell based on dye-sensitized colloidal TiO₂ films", *Nature*, vol. 353, pp. 737–740, October 1991.
- [2] M. Grätzel, "Recent advances in sensitized mesoscopic solar cells", *Acc. Chem. Res.* vol. 42, pp. 1788–1798, August 2009.
- [3] H. Wang, P. Nicholson, L. Peter, S. Zafeeruddin and M. Grätzel, "Transport and interfacial transfer of electrons in dye-sensitized solar cells utilizing a co(dbbip)₂ redox shuttle", *J. Phys. Chem. C.*, vol.114, pp. 14300-14306, August 2010.
- [4] K. H. Park, and M. Dhayal, "High efficiency solar cell based on dye sensitized plasma treated nanostructured TiO₂ films", *Electrochemistry Communications*, vol. 11, pp.75-79, January 2009.
- [5] S. A. Taya, T. M. El-Agez, H. S. El-Ghamri and M. S. Abdel-Latif, "Dye-sensitized solar cells using fresh and dried natural dyes", *International Journal of Materials Science and Applications*, vol. 2, pp. 37-42, March 2013.
- [6] T. M. El-Agez, A. A. El Tayyan, A. Al-Kahlout, S. A. Taya and M. S. Abdel-Latif, "Dye-sensitized solar cells based on ZnO films and natural dyes", *International Journal of Materials and Chemistry*, vol. 2, pp. 105-110, August 2012.
- [7] M. S. Abdel-Latif, T. M. El-Agez, S. A. Taya, A. Y. Batniji and H. S. El-Ghamri, "Plant Seeds-Based Dye-Sensitized Solar Cells", *Journal of Materials Sciences and Applications*, vol. 4, pp. 516-520, September 2013.
- [8] G. Calogero and G. Di. Marco, "Red Sicilian orange and purple eggplant fruits as natural sensitizers for dye-sensitized solar cells", *Sol. Energ. Mat. Sol. C.*, vol. 92, pp. 1341–1346, November 2008.
- [9] K. Wongcharee, V. Meeyoo and S. Chavadej, "Dye-sensitized solar cell using natural dyes extracted from rosella and blue pea flowers", *Sol. Energ. Mat. Sol. C.*, vol. 91, pp. 566–571, April 2007.
- [10] M.S. Roy, P. Balraju, M. Kumar and G.D. Sharma, "Dye-sensitized solar cell based on Rose Bengal dye and nanocrystalline TiO₂", *Sol. Energ. Mat. Sol. C.* vol. 92, pp. 909–913, August 2008.
- [11] Z. Wang, Y. Cui, K. Hara, Y. Dan-oh, C. Kasada and A. Shinpo, "A high-light harvesting- efficiency coumarin dye for stable dye-sensitized solar cells", *Adv. Mater.*, vol. 19, pp. 1138-1141, April 2007.
- [12] J. Etula, "Comparison of three Finnish berries as sensitizers in a dye-sensitized solar cell", *European Journal For Young Scientists And Engineers*, vol. 1, pp. 5-23, November 2012.
- [13] R. Ahmadian, "Estimating the impact of dye concentration on the photoelectrochemical performance of anthocyanin-sensitized solar cells: a power law model", *Journal of photonics for energy*, vol. 1, pp. 12-15, January 2011.
- [14] A. R. Hernández-Martínez, M. Estevez, S. Vargas, F. Quintanilla and R. Rodríguez, "Natural Pigment-Based Dye-Sensitized Solar Cells", *1st international congress on instrumentations and applied sciences*, vol. 10, pp. 1665-6423, February 2012.
- [15] A. Y. Batniji, R. Morjan, M. S. Abdel-Latif, T. M. El-Agez, S. A. Taya, and Hatem S. El-Ghamri, "Aldimine derivatives as photosensitizers for dye-sensitized solar cells", *Turkish Journal of Physics*, vol. 38, pp. 86–90, January 2014.
- [16] A. Bakowska, A. Z. Kucharska and J. Oszmianski, "The effects of heating, UV irradiation, and storage on stability of the anthocyanin-polyphenol copigment complex", *Food Chem.*, vol. 81, pp. 349-355, June 2003