Solar Cells Sensitized with the Extracts of Hibiscus Sabdariffa and Rosa Damascena Flowers

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Abstract- Dye sensitized solar cells (DSSCs) based on natural dyes extracted from dried Hibiscus Sabdariffa and Rosa Damascena flowers were fabricated. The extracts were characterized by UV-Vis absorption spectroscopy. The TiO_2 nanopowder paste was spread on FTO layers to form a thin layer. Three processes were conducted to improve the cell efficiency such as pre- and post-treatments of TiO_2 and FTO layers with three acids and changing the pH of the extract solutions. The conversion efficiency of the DSSCs sensitized with Hibiscus Sabdariffa enhanced by 107% and 114% when treating the FTO sheets for 5 min with H_3PO_4 and HCl, respectively. It was observed that when HCl acid was used to adjust the pH value of the dye solution of Rosa Damascena to 2.0, the efficiency of the DSSC was enhanced by about 180%.

Keywords: dye sensitized solar cells, natural dyes, Hibiscus Sabdariffa, Rosa Damascena, TiO_2.

1. Introduction

Dye-sensitized solar cell (DSSC) is a photoelectrochemical device that converts visible light into electrical energy. DSSC is one of the third generation of solar cells. In 1991, DSSC was developed by B. O'Regan and M. Grätzel with energy conversion efficiency of 7% [1]. Since then DSSCs have received an increasing interest due to many features such as low-cost, environmentally friendly, and simple fabrication techniques. A DSSC has a simple structure based on a nanocrystalline wide band-gap metal oxide semiconductor (e.g. TiO_2) coated on a fluorine-doped tin oxide (FTO) layer, dye adsorbed on the semiconducting material film, redox electrolyte solution (e.g. I^-/I^{-3}) sandwiched between a counter electrode and the dye molecule, and counter electrode as a thin film of a platinum (Pt). The principal of operation is based on the sensitization of a wide band-gap metal oxide semiconductor to visible light region by the adsorbed dye molecules. The dye molecules become excited by absorbing photons from the incident sun light. The excited electrons are injected into the conduction band of the metal oxide layer and the dye becomes oxidized. The excited dye is then reduced by the electron donor from the electrolyte. The excited electrons flow through the external load circuit toward the platinum electrode which regenerates the redox electrolyte.

The sensitizing dye plays a crucial role in the DSSC operation. Natural dyes have received a considerable interest in the fabrication of DSSCs because they are inexpensive, nontoxic, and widely available. Several papers have been published on using natural dyes as photosensitizers of DSSCs [2-16]. Among these natural dyes pomegranate juice [2], black grapes [3], red amaranth [4], red turnip [5], hibiscus [6], chlorophyll [7, 8], dried plant leaves [9-11], plant seeds [12], and others [13] have been investigated as sensitizers of dye-sensitized solar cells.

In this work, two dyes were extracted from dried coronet of Hibiscus Sabdariffa and Rosa Damascena flowers and used as photosensitizers of DSSCs. The extracts were characterized by UV-Vis absorption spectra. The TiO_2 nanopowder was used as a semiconductor. The current-voltage characteristic curves of the fabricated cells were measured and the photoelectrochemical parameters were calculated. The effects of acidic treatment of TiO_2 layer and FTO sheet on the cell performance were examined. Moreover, the effect of pH of dye solution on the DSSC efficiency was also investigated.

2. Related studies

DSSCs were assembled using TiO_2 electrode and synthesized with the extract of red amaranth. Water, ethanol, methanol and acetone were used as solvents of the dye extract. The highest efficiency of 0.22% was obtained when the sensitization time of electrode was 30 min and dye was extracted by acetone [4]. The extracts of bougainvillea flowers, red turnip and purple wild Sicilian prickly pear fruit juice were used as sensitizers of TiO_2 electrode [5]. The best light to electricity conversion efficiency of 1.7% was obtained, under AM 1.5 irradiation, with the red turnip extract. The extract of green cabbage was tested as a sensitizer and produced a photoelectric conversion efficiency of 0.1%, an open-circuit voltage of 532 mV, and a short-circuit current density of 1.2 mA/cm^2 [8]. DSSCs based on natural dyes extracted from six dried plant leaves were fabricated [9]. The best performance was obtained for the DSSC sensitized with Jasminum Grandifolium with an efficiency of 0.335%. Natural dyes were extracted from dried plant leaves of Cream, Apricot, Figs, Apples, Sage, Thyme, Mint, Zizyphus jujube, Orange, Shade tree, Basil, Berries, Mirabelle plums, Victoria plums, Peach, Mango, Pomegranate, Bananas, Guava, and Fluoridation and tested for the sensitization of TiO_2 films [10]. The best performance was observed for the DSSC sensitized with Zizyphus jujube. Preparation procedures and optical and electrical characterization of DSSCs sensitized by Henna, pomegranate, cherries and Bahraini raspberries were investigated [17]. The authors found that these natural dyes are potential candidates to replace some of the synthetic dyes. Gold nanoparticles as a Schottky barrier were fabricated on a TiO_2 layer to assemble a DSSC which was synthesized using extracts of the natural dye Rhoeo spathacea [18]. The extracts of rosella, blue pea, and mixed of them were investigated as photosensitizers of DSSCs [19]. The light absorption spectrum of the mixed extract contained peaks corresponding to the contributions from both rosella and blue pea extracts. The energy conversion efficiencies were 0.37%, 0.05% and 0.15%, for the DSSCs sensitized with rosella extract alone, blue pea extract alone and mixed extract, respectively. A food pigment (Monascus yellow) extracted from Monascus fermentations (red yeast rice) was used as sensitizer of DSSCs [20]. The DSSC fabrication process has been optimized in terms of the rinsing solvent used after dye adsorption and the dye-uptake duration. DSSCs were assembled using the extracts of black rice, capsicum, erythrina variegata flower, rosa xanthina, and kelp [21]. The short circuit current was found to range from 0.225 mA to 1.142 mA, the open circuit voltage took the range from 412 mV to 551 mV, and the fill factor had the range from 0.52 to 0.63.

3. Experimental work

The dried coronets of Hibiscus Sabdariffa and Rosa Damascena flowers were used as sensitizers of the fabricated DSSCs. They were washed by distilled water and dried at 60 ºC. 15 g of the dried flowers were immersed in 10 ml of ethyl alcohol at room temperature and in dark for 24 hr. The extracts of the natural dyes were obtained after filtration of the solutions.

FTO conductive glass sheets of resistance of 20 Ω/cm^2 and transmission >80% (Xinyan Technology Ltd., Hong Kong) were used as substrates. The substrates were cleaned in a detergent solution using an ultrasonic bath for 20 min twice, then immersed in boiling water for 30 min. They were dried and kept in an oven at 60 ºC. The TiO_2 paste was prepared by adding 2 gm of TiO_2 nanopowder of 10-25 nm size, 4 ml distilled water, 10 µl acetyl acetone and 50 µl Triton X-100. The mixture was grinded for half an hour until a homogeneous paste was obtained. Doctor blade method was used to distribute the TiO_2 paste on the FTO glass sheets. Two pieces of tape with thickness of 20 µm were used to restrict the TiO_2 film thickness and area. The active area of the film was 0.25 cm^2. The films were sintered at 450 ºC for 30 min and then were cooled down to about 100 ºC. They were dyed for 24 hr under dark. The dyed TiO_2 electrode and platinum counter electrode were assembled to form a solar cell by sandwiching a redox (I^-/I^{-3}) electrolyte solution.

Three treatments were conducted, namely treating the FTO and TiO_2 layers with three acids and changing the pH of the dye solutions using acetic and hydrochloric acids. In the first treatment, the cleaned FTO substrates were immersed in diluted hydrochloric, nitric, or phosphoric acid of 0.1 M for 5 min. In order to examine the effect of immersion time, another group of FTO sheets were immersed in the acids for 10 min. In the second treatment, the sintered TiO_2 layers were immersed in diluted hydrochloric, nitric, or phosphoric acids of 0.1 M for 5 min. In the third treatment, a 0.1 M concentration of hydrochloric and acetic acids were added to the dye solutions to change the solution pH value. Four and three different pH values ranging from 5.5 to 2.0 and 1.5 to 3 were examined for the dye solutions of Rosa Damascena and Hibiscus Sabdariffa, respectively.

The absorption spectra measurements of the extracted dyes of Hibiscus Sabdariffa and Rosa Damascena flowers in ethyl alcohol solution were performed using Genesys 10S UV-Vis spectrophotometer with code L9M3130012 and a range of wavelength from 400 nm to 800 nm was adopted. The J-V characteristic curves of the fabricated DSSCs were measured under an illumination of 100 mW/cm^2 using National Instruments data acquisition card (USB NI 6251) in combination with Lab View program with a high pressure mercury arc lamp with IR filter.

4. Results and discussions

4.1. Absorption spectra of natural dyes

Figure 1 shows the UV-Vis absorption spectra of the natural extracts of Hibiscus Sabdariffa and Rosa Damascena flowers. The figure shows the presence of distinct absorption peaks in the visible region. The absorption peaks of Hibiscus Sabdariffa dye can be seen at about 545 nm and 664 nm. The absorption peak of Rosa Damascena dye is at 541.63 nm.

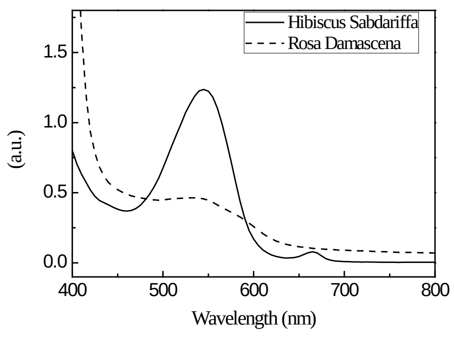


Fig. 1. The UV-Vis absorption spectra of the dyes extracted from Hibiscus Sabdariffa and Rosa Damascena.

4.2. Photoelectrochemical Properties

The J-V measurements of the DSSCs sensitized with the extracts of Hibiscus Sabdariffa and Rosa Damascena flowers are illustrated in Fig. 2. These values were measured under an illumination of 100 mW/cm^2. To investigate the DSSC performance, short circuit current density, Jsc, open circuit voltage, V_{oc}, maximum power point, fill factor, FF, and conversion efficiency, η, must be determined. Short circuit current density and open circuit voltage can be determined from the J-V curve interceptions with y- and x-axes, respectively. The DSSC output power can be calculated as P = JV. From the output power data, the maximum power point can be obtained. Using these parameters and the input power, fill factor and conversion efficiency can be calculated. All photoelectrochemical parameters of the fabricated DSSCs are presented in Table 1. V_m and I_m in Table 1 represent the voltage and current of maximum power point. V_{oc} was found to be 0.46 V for the DSSC sensitized with Hibiscus Sabdariffa extract and 0.53 V for that dyed with Rosa Damascena. The J_{sc} has a value of 1.18 mA/cm^2 for the DSSC sensitized with the dye extracted from Hibiscus Sabdariffa and 0.35 mA/cm^2 for the DSSC sensitized with the dye extracted from Rosa Damascena. The fill factor was found to be 51% and 56% for Hibiscus Sabdariffa and Rosa Damascena, respectively. The efficiencies were 0.28% for the DSSC sensitized with Hibiscus Sabdariffa and 0.11% for that sensitized with Rosa Damascena.

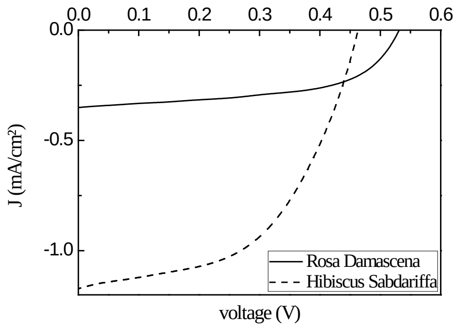


Fig. 2. Current density-voltage characteristic curves for the DSSCs sensitized with Hibiscus Sabdariffa and Rosa Damascena.

Table 1. Photovoltaic parameters of the DSSCs sensitized by Hibiscus Sabdariffa and Rosa Damascena.

Dye	V_{oc} (V)	J_{sc} (mA/cm²)	V_m (V)	J_m (mA/cm²)	FF	η %
Hibiscus Sabdariffa	0.46	1.18	0.32	0.89	0.51	0.28
Rosa Damascena	0.53	0.35	0.41	0.26	0.56	0.11

4.3. FTO acidic treatment

Figure 3 shows the J-V characteristics of DSSCs fabricated using the treated FTO with H_3PO_4, HCl, or HNO_3 acids for 5 min (upper panels) and 10 min (lower panels). The photovoltaic parameters of these DSSCs are listed in Table 2. The V_{oc} ranges between 0.45 V and 0.53 V. The J_{sc} has a maximum value of 1.20 mA/cm^2 for the DSSC sensitized with the Hibiscus Sabdariffa dye when treating FTO with H_3PO_4 acid for 5 min, and a minimum value of 0.23 mA/cm^2 for the DSSC sensitized with the Rosa Damascena dye when treating FTO with H_3PO_4 acid for 10 min. The highest fill factor (57%) was obtained for the DSSC sensitized with the extract of Hibiscus Sabdariffa when treating FTO with HCl acid for 5 min and the lowest fill factor (45%) was observed for the cell dyed with Rosa Damascena when treating FTO with H_3PO_4 acid for 10 min. The highest efficiency of 0.32% was obtained for the DSSC sensitized with Hibiscus Sabdariffa when treating FTO with HCl acid for 5 min whereas the lowest efficiency of 0.05% was observed for the DSSC sensitized with Rosa Damascena when treating FTO with H_3PO_4 acid for 10 min. The treatment of FTO layer by the three acids didn't improve the efficiency of DSSCs sensitized with Rosa Damascena. The fill factor of the DSSCs sensitized with Hibiscus Sabdariffa enhanced by 106% and 117% when treating the FTO sheets for 5 min with H_3PO_4 and HCl, respectively whereas the efficiencies of these cells improved by 107% and 114% for these acids.

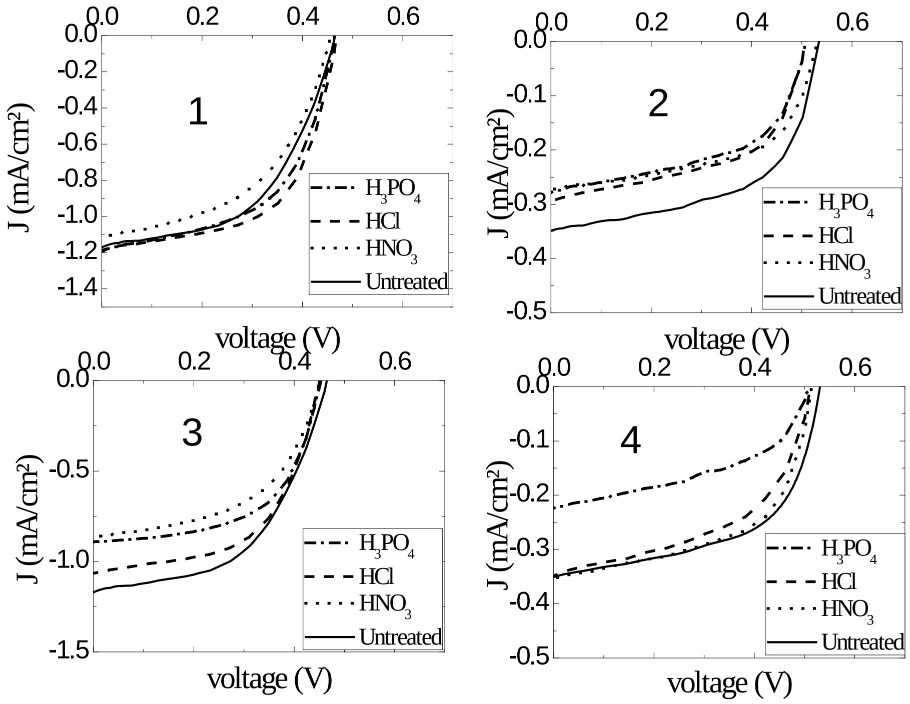


Fig. 3. Current density-voltage characteristic curves of the DSSCs fabricated using the pre-treatment of FTO layer with one of three acids. The DSSCs are sensitized with Hibiscus Sabdariffa (1 and 3) and Rosa Damascena (2 and 4). Upper and lower panels present the 5 min and 10 min treatment, respectively.

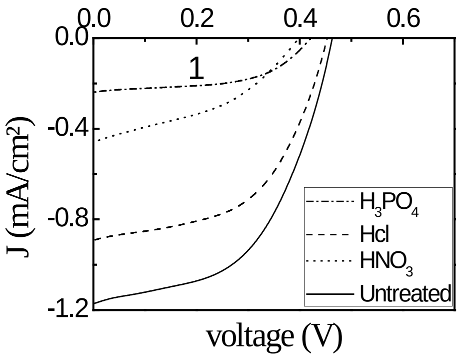
Table 2. Photovoltaic parameters of the DSSCs using the treated FTO with the acids for 5 and 10 min.

Dye	Time	Acids	V_{oc} (V)	J_{sc} (mA/cm²)	V_m (V)	J_m (mA/cm²)	FF	η %
Hibiscus Sabdariffa	Untreated		0.46	1.18	0.32	0.89	0.51	0.28
	5 Min	H_3PO_4	0.46	1.20	0.34	0.87	0.54	0.30
		HCl	0.47	1.19	0.35	0.92	0.57	0.32
		HNO_3	0.46	1.13	0.32	0.78	0.48	0.25
	10 min	H_3PO_4	0.46	0.87	0.34	0.65	0.55	0.22
		HCl	0.45	1.07	0.33	0.82	0.56	0.27
		HNO_3	0.45	0.88	0.33	0.61	0.51	0.20
Rosa Damascena	Untreated		0.53	0.35	0.41	0.26	0.56	0.11
	5 min	H_3PO_4	0.51	0.28	0.40	0.19	0.52	0.08
		HCl	0.51	0.29	0.40	0.20	0.54	0.08
		HNO_3	0.53	0.28	0.42	0.19	0.54	0.08
	10 min	H_3PO_4	0.51	0.23	0.38	0.14	0.45	0.05
		HCl	0.51	0.34	0.38	0.22	0.48	0.08
		HNO_3	0.51	0.36	0.40	0.25	0.54	0.10

4.4. Acidic treatment of TiO_2

Figure 4 illustrates the J-V characteristics of the DSSCs fabricated using the treated TiO_2 film with one of the three acids for 5 min. We here call the TiO_2 acid treatment as post treatment. Table 3 presents the photovoltaic parameters of the TiO_2-treated DSSCs using H_3PO_4, HCl, or HNO_3 acids. The V_{oc} ranges between 0.40 V for the DSSC sensitized with Hibiscus Sabdariffa and post treated with HNO_3 acid and 0.53 V for the untreated cell dyed with Rosa Damascena and for that post-treated with HCl acid. The J_{sc} has a maximum value of 1.18 mA/cm^2 for the untreated DSSC sensitized with the dye extracted from Hibiscus Sabdariffa. The fill factor of the fabricated cells changed from 39% to 58%. The highest fill factor was obtained for the DSSC sensitized with the extract of Rosa Damascena using treated TiO_2 film with H_3PO_4 acid and the lowest fill factor was observed for the cell dyed with Hibiscus Sabdariffa using treated TiO_2 film with HNO_3 acid. The highest efficiency has a value of 0.28% for the untreated DSSC sensitized with Hibiscus Sabdariffa whereas the lowest efficiency has a value of 0.05% for the DSSC sensitized with Hibiscus Sabdariffa when treating TiO_2 layer with H_3PO_4 acid. It is clear that the post-treatment of TiO_2 by the three acids for 5 min didn't improve the output power and efficiency when the Rosa Damascena and Hibiscus Sabdariffa dye solutions were used as photosensitizers.

The reasons behind the decline in the photovoltaic parameters after the post treatment of TiO_2 film may be due to the low amount of dye adsorbed onto the TiO_2 film and poor connection between material interfaces.



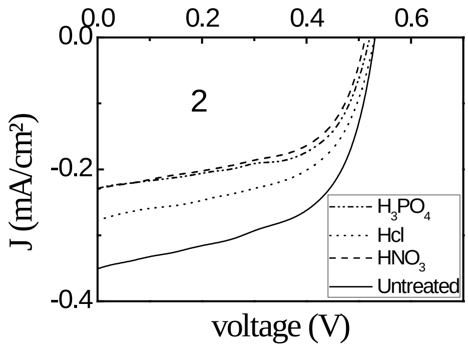


Fig. 4. Current density-voltage characteristic curves of the DSSCs sensitized by (1) Hibiscus Sabdariffa and (2) Rosa Damascena with post-treated TiO_2 film by the acids for 5 min.

Table 3. Photovoltaic parameters of the DSSCs fabricated using post-treated TiO_2 film by the three acids for 5 min.

Dye	Acids	V_{oc} (V)	J_{sc} (mA/cm²)	V_m (V)	J_m (mA/cm²)	FF	η %
Hibiscus Sabdariffa	Untreated	0.46	1.18	0.32	0.89	0.51	0.28
	H_3PO_4	0.43	0.23	0.32	0.15	0.48	0.05
	HCl	0.45	0.90	0.32	0.66	0.52	0.21
	HNO_3	0.40	0.46	0.26	0.28	0.39	0.07
Rosa Damascena	Untreated	0.53	0.35	0.41	0.26	0.56	0.11
	H_3PO_4	0.52	0.23	0.41	0.17	0.58	0.07
	HCl	0.53	0.28	0.42	0.20	0.56	0.08
	HNO_3	0.52	0.23	0.38	0.16	0.50	0.06

4.5. Effect of the dye solution pH

The J-V characteristics of the fabricated DSSCs with different pH values of the dye solutions are shown in Fig. 5. Table 4 presents the photovoltaic parameters of these DSSCs. The pH values of the dye solutions were varied using acetic and hydrochloric acid. As can be seen in the Table, V_{oc} ranged between 0.38V for the DSSC dyed with the extract of Hibiscus Sabdariffa of pH of 1.5 adjusted with HCl acid to 0.53V for the DSSC sensitized with untreated Rosa Damascena. The J_{sc} has a maximum value of 1.18 mA/cm^2 for the DSSC dyed with untreated Hibiscus Sabdariffa and a minimum value of 0.17 mA/cm^2 for that sensitized with Hibiscus Sabdariffa of pH of 1.5 adjusted with HCl acid. The fill factor of the fabricated cells varied from 42% to 58%. The highest efficiency of 0.28% was obtained for the DSSC dyed of Hibiscus Sabdariffa at initial pH before the addition of the acids to the dye solution. The acetic acid treated dyes showed poor performance in the power conversion efficiency which may be due to changing of the molecular structure of the dye. Fig. 6 shows the DSSC efficiency as a function of the extract solution pH using acetic and hydrochloric acids. In general, the efficiency is diminished with decreasing the pH for all cells dyed with Hibiscus Sabdariffa and Rosa Damascena when using acetic and hydrochloric acid to change the dye solution pH. Only the efficiency of the DSSC sensitized with Rosa Damascena extract with pH of 2.0, adjusted using HCl acid, was dramatically enhanced. An efficiency improvement of 180% was obtained. The reason of the general decline in the conversion efficiency of the DSSCs with varying the pH value of the dye solutions may be attributed to the poor bonding between dye molecules and TiO_2 film and this leads to slow the electron injection from dye excited state into conduction band of TiO_2. Moreover, the variation of pH value can decrease the optical absorption of the dye.

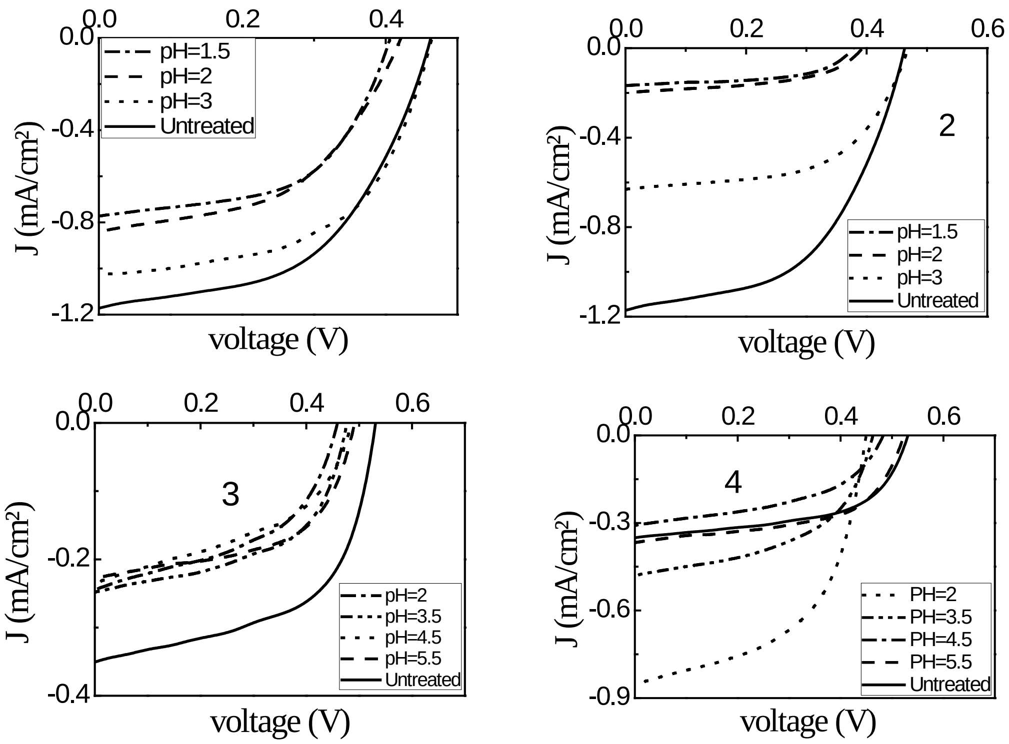


Fig. 5. Current density-voltage characteristic curves for the DSSCs sensitized by (1) Hibiscus Sabdariffa and (3) Rosa Damascena treated with acetic acid, and those sensitized by (2) Hibiscus Sabdariffa and (4) Rosa Damascena treated with hydrochloric acid.

Table 4. Photovoltaic parameters of the DSSCs sensitized by natural dye solutions having different pH values.

Dye	Acid	pH	V_{oc} (V)	J_{sc} (mA/cm²)	V_m (V)	J_m (mA/cm²)	FF	η %
Hibiscus Sabdariffa	untreated	3.23	0.46	1.18	0.32	0.89	0.51	0.28
	acetic acid	1.5	0.40	0.77	0.28	0.61	0.55	0.17
		2.0	0.42	0.84	0.28	0.60	0.47	0.17
		3.0	0.46	1.03	0.34	0.76	0.54	0.26
	HCl Acid	1.5	0.38	0.17	0.30	0.11	0.51	0.03
		2.0	0.39	0.20	0.30	0.13	0.50	0.04
		3.0	0.47	0.63	0.35	0.49	0.58	0.17
Rosa Damascena	Untreated	6.15	0.53	0.35	0.41	0.26	0.56	0.11
	acetic acid	2.0	0.46	0.24	0.34	0.16	0.49	0.05
		3.5	0.48	0.25	0.37	0.17	0.53	0.06
		4.5	0.49	0.25	0.37	0.14	0.42	0.05
		5.5	0.49	0.23	0.38	0.16	0.54	0.06
	HCl Acid	2.0	0.45	0.85	0.34	0.59	0.52	0.20
		3.5	0.46	0.48	0.36	0.31	0.50	0.11
		4.5	0.48	0.31	0.36	0.20	0.48	0.07
		5.5	0.52	0.37	0.41	0.26	0.55	0.11

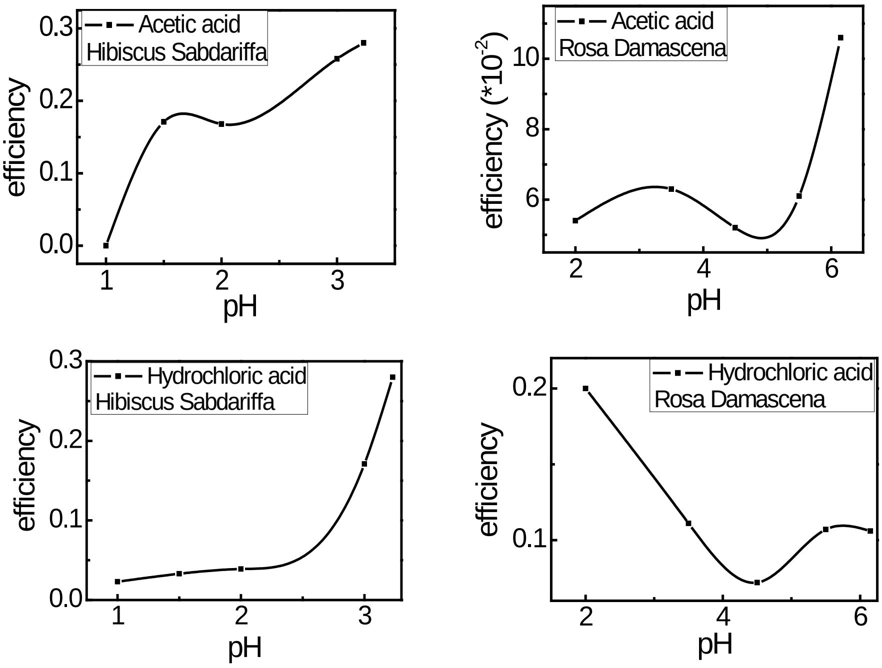


Fig. 6. DSSC efficiency versus the pH of the extract solution using acetic and hydrochloric acid**.**

5. Conclusions

DSSCs were fabricated using TiO_2 as a semiconducting material. Natural dyes extracted from Hibiscus Sabdariffa and Rosa Damascena flowers were used as photosensitizers. The absorption spectra of these dyes and the J-V characteristics of the DSSCs were performed. The photovoltaic parameters of the fabricated DSSCs were calculated. The best performance was obtained for the DSSC sensitized with Hibiscus Sabdariffa with V_{oc} = 0.46 V, J_{sc} = 1.18 mA/cm^2, V_m = 0.32 V, J_m = 0.89 mA/cm^2, FF = 0.51 and η = 0.28. Different treatments were examined such as acid treatment of TiO_2 and FTO and changing the pH of dye solutions. The fill factor of the DSSCs sensitized with Hibiscus Sabdariffa enhanced by 106% and 117% when treating the FTO sheets for 5 min with H_3PO_4 and HCl, respectively. Moreover, the conversion efficiency of these cells was also improved by 107% and 114% when treating the FTO sheets for 5 min with H_3PO_4 and HCl, respectively. The post-treatment of TiO_2 by H_3PO_4, HCl, and HNO_3 acids for 5 min didn't show any efficiency improvement. When HCl acid was used to adjust the pH value of the dye solution of Rosa Damascena to 2.0, the efficiency of the DSSC was enhanced by about 180%.

In comparison of the current work with similar studies utilizing natural dyes, we find that the efficiencies obtained in the current work with the extract of Hibiscus Sabdariffa are higher than those obtained by J. Uddin et al. [4], L. Amadi et al. [8], and S. Hao et al. [21] and they are almost the same as those obtained by S. Taya et al. [9] and K. Wongchareea et al. [19].

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