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Performance of Purple Carrot as a Sensitizer for Dye-sensitized Solar Cells

[1]Islam M. Radwan, [1,2]Taher M. El-Agez, [1,2]Sofyan A. Taya, [2,3]Monzir S. Abdel-Latif, [1,2]Hatem S. Ghamri, [4]Amal M. Al-Kahlout and [4]Naji Al Dahoudi

[1]Physics Department, Islamic University of Gaza, Gaza, Palestine.
[2]Renewable Energy Center, Islamic University of Gaza, Gaza, Palestine.
[3]Chemistry Department, Islamic University of Gaza, Gaza, Palestine.
[4]Physics Department, Al Azhar University, Gaza, Palestine.

Abstract: Dye-sensitized solar cells (DSSCs) have been the subject of intensive research during the last few years due to their low production and fabrication costs. In this work, DSSCs were prepared using titanium dioxide (TiO_2) as a semiconducting layer and sensitized using three natural dyes extracted from plant roots, such as, purple carrot, beet and curcuma. Among these dyes, purple carrot showed an efficiency of 0.32%, which was the highest. The performance of DSSCs sensitized by purple carrot was investigated with the dye extraction temperature and pH of the extract solution. The results showed that an extracting temperature of 323.15 °K could be used as an optimal value. The treatment of dye solutions with acetic acid to adjust the pH to 2.8 resulted in an efficiency of 0.6%.

Key words: Dye-sensitized solar cells, Purple carrot, pH.

INTRODUCTION

The tremendous increase in energy consumption in the last few decades and the exhausted fossil fuels supply led to an urgent need to find other substitutional energy resources to meet the ever increasing energy demand. Solar energy is considered one of the most promising energy sources for the future. Dye-sensitized solar cells (DSSCs) have been investigated intensively during the last twenty years, due to their low production and fabrication costs (Taya *et al*., 2014; Batniji *et al*., 2014; Abdel-Latif *et al*., 2015; Taya *et al*., 2015; El-Agez *et al*., 2014; El-Ghamri *et al*., 2015). A DSSC belongs to the third generation photovoltaic cells that uses molecules to absorb photons and separates the two functions of light harvesting and charge transport. These cells have the benefits of suitability for many materials, and their possibility of production, under mild conditions, makes them significantly less expensive than the earlier cell designs. The breakthrough in the fabrication of DSSCs occurred in 1991, when O'Regan and Grätzel, at Ecole Polytechnique Federale de Lausanne (EPEL) in Switzerland, used TiO_2 nanoparticles to expand the surface area available for dye adsorption (O'Regan and Gratzel, 1991).

A DSSC is generally composed of a mesoporous metal oxide semiconductor (usually TiO_2) coated on transparent conducting glass (fluorine doped tin oxide FTO) and a sensitizer adsorbed onto the surface of the semiconductor acting as antennae to harvest as much as possible of the solar spectrum reaching the Earth's surface. Finally, the sensitized mesoporous film is capped with a platinum counter electrode while the in-between space is filled with a liquid electrolyte containing suitable redox couples (e.g., I_3^-/I^-).

The operating cycle of DSSCs starts from the excitation of sensitizing dyes by light absorption followed by electron transfer from the dye's excited state into the conduction band of TiO_2. Electron transports through the mesoporous network of TiO_2 particles by diffusion reaching the FTO. The electrolyte solution reduces the oxidized dye molecules and, at the same time, the electrolyte is regenerated by the electrons injected from the counter electrode (Hauch and Georg, 2001). Many metal complexes and organic dyes have been used as sensitizers. Ruthenium (Ru) complexes are one of the best sensitizers for DSSCs because of their wide band absorbance and highly efficient metal-to-ligand charge transfer (Gratzel, 2009). On the other hand, rarity, high cost and the complicated synthesis of ruthenium complexes are their main disadvantages. Significant research interest has developed in finding alternative, readily available and low cost efficient photosensitizers. Recently, enormous easily available dyes extracted from natural sources as photosensitizers are under focus because of

Corresponding Author: Sofyan A. Taya, Physics Department/Renewable Energy Center,, Islamic University of Gaza, Gaza, Palestine.
E-mail: staya@iugaza.edu.ps

their large absorption coefficients, high light-harvesting efficiency, low cost, easy preparation and environment friendliness.

In this work, natural dyes, extracted from purple carrot, beet and curcuma, have been tested as photosensitizers of DSSCs. The absorbance spectra of these dyes were examined, in addition to the photovoltaic properties of the fabricated cells. Moreover, the performance of DSSCs sensitized by purple carrot was investigated with the dye extraction temperature and pH of the extract solution. Finally, the electrochemical impedance spectroscopy was performed for a DSSC sensitized by purple carrot of pH 2.8.

MATERIALS AND METHODS

Three types of plant roots were collected from purple carrot, beet, and curcuma. These roots were washed intensively with water to remove the adhering particles and dust from their surfaces and then dried. One gram of each root was immersed in 10 ml of ethanol at room temperature and kept in the dark for one day. After filtration of the solutions, natural extracts were obtained.

Fluorine doped tin oxide (FTO) glass substrates, each having 15-20 Ω/□ resistance, were cleaned by acetone and water, successively for 20 minutes using an ultrasonic bath and then dried. The semiconductor paste was prepared by blending 2 g of TiO_2 (Sigma Aldrirch, 97%) nanopowder, 4 ml of distilled water, 10 µl of acetylacetone (Sigma Aldrirch, 99.8%), and 50 µl of Triton X-100 (Fluka-Analytical, 99%) as the surfactant (Hara *et al*., 2000). The TiO_2 paste was spread uniformly on the substrate by doctor-blade technique in order to obtain a TiO_2 thin film about 20µm thickness. The FTO/TiO_2 film was sintered to 723.15 °K for half an hour, and then the TiO_2 deposited- electrode was cooled down to 100 °C. The FTO/TiO_2 electrodes were immersed in the natural extracts for 24 h. Finally, all parts of the DSSCs were assembled by fixing the dyed TiO_2 electrode and the Pt counter electrode so that the dyed TiO_2 plates facing down onto the coated Pt counter electrode. The two electrodes were pressed and clamped firmly in a sandwich configuration. The electrolyte redox (I^-/I_3^-) (Himedia, 99.9%) was spread between the two electrodes, using micropipette. The fabricated cells were exposed to light in order to study their photovoltaic properties.

To investigate the effect of the extraction temperature of the purple carrot dye, one gram of purple carrot was immersed in 10 ml of ethanol at five different extracting temperatures, 298.15 °K, 313.15 °K, 323.15 °K, 333.15 °K and 343.15 °K, using an oven and were kept in the dark for one day. The fabricated TiO_2 films were soaked in these different dyes solutions for 24 h. After assembling the DSSCs, the photovoltaic parameters were studied.

To investigate the effect of pH on the performance of purple carrot extracts as photosensitizers, dyes were treated using acetic and hydrochloric acids. The original pH of the purple carrot extracts in ethanol was about 7.4. Five different pH values, ranging from 2.8 to 7.4, were examined by adding different amounts of (0.1 M) acetic acid to the dye solutions. On the other hand, the same procedure was applied using 0.1 M of hydrochloric acid with different amounts added to the dye solutions in order to get six different values of pH ranging from 7.4 to a minimum value of 1. Finally the photovoltaic properties of the cells were investigated.

RESULTS AND DISCUSSION

Absorption of natural dyes: The absorption spectra of three dye solutions of purple carrot, beet and curcuma were investigated, using GENESYS 10S UV-Vis spectrophotometer. The absorption spectra were recorded in ethanol as solvent in the wavelength range from 390 to 700 nm. Fig. 1 shows the UV–Vis absorption spectra of three natural dyes extracted from three plant roots. It is clear from Fig. 1, that purple carrot has absorbance peaks at 552 nm and 427 nm. For the extract of beet root, it has an absorption peak at 544 nm, while extract of curcuma has an absorption peak at 424 nm.

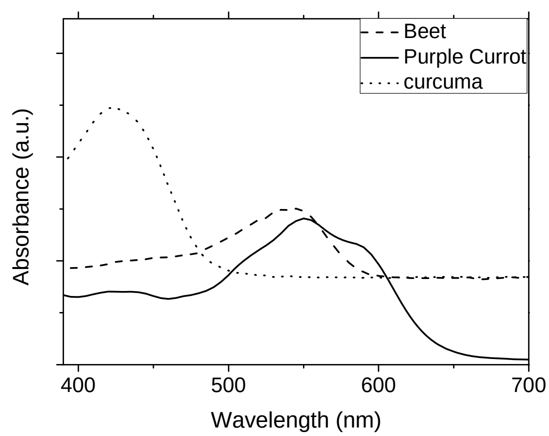

Fig. 1: UV–Vis absorption spectra of beet, purple carrot, and curcuma.

It may be observed from Fig. 1 that purple carrot has a wide band of absorption. This means that it is a good candidate for sensitization. This can be attributed to the existence of anthocyanin pigment (Cevallos-Casals and Cisneros-Zeballos, 2004).

J-V characterization of DSSCs: The current density (J) - voltage (V) curves of the fabricated DSSCs are illustrated in Fig. 2. Short circuit current (J_{sc}) and open circuit voltage (V_{oc}) can be obtained from the J-V characteristic curves. The photovoltaic parameters, such as, maximum current density (J_m), maximum voltage (V_m), fill factor (FF), and efficiency (η) of the DSSCs, are listed in Table 1. The fill factor of the cell can be calculated by equation 1.

$$FF = \frac{V_m \ I_m}{V_{oc} \ I_{sc}} \qquad \ldots.. \qquad (1)$$

where V_m is the optimum point voltage, I_m is the optimum point current, V_{oc} is the open circuit voltage and I_{sc} is the short circuit current.

The overall cell efficiency can be calculated by equation 2.

$$\eta_{eff} = \frac{V_{oc} \ I_{sc} \ FF}{P_{in}} = \frac{V_m \ I_m}{P_{in}} \qquad \ldots \qquad (2)$$

where P_{in} is the power input from the sunlight.

Any inspection of Table 1 shows that the best performance was obtained with DSSC with the extract of purple carrot. This cell was η = 0.32%, whereas, the lowest performance was with that sensitized with beet where η = 0.05%.

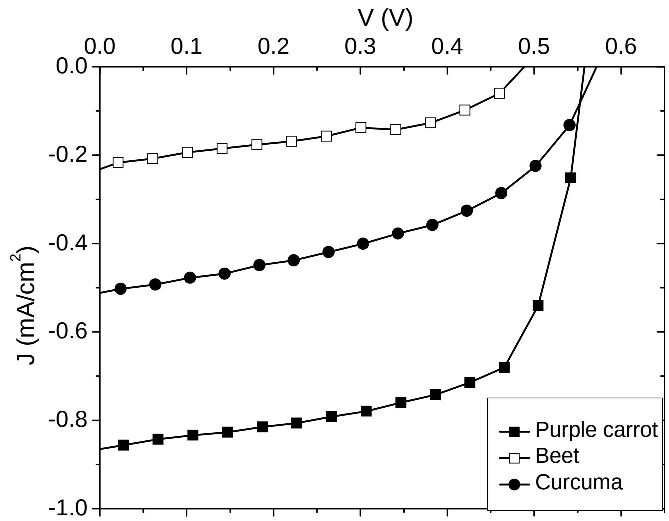

Fig. 2: J-V characteristic curves for DSSCs sensitized by the extracts of purple carrot, beet, and curcuma.

Table 1: Photovoltaic parameters of DSSCs sensitized by natural dyes extracted from plant roots compared with that sensitized with Ru (N719).

Dye	J_{sc} (mA/cm^2)	V_{oc} (V)	J_m (mA/cm^2)	V_m (V)	FF	η %
Purple carrot	0.87	0.56	0.69	0.46	0.66	0.32
Curcuma	0.51	0.57	0.33	0.42	0.47	0.14
Beet	0.23	0.49	0.14	0.36	0.43	0.05
Ru (N719)	4.76	0.57	3.53	0.38	0.50	1.35

Purple carrot exhibits the highest performance, so the next studies will be conducted with DSSC sensitized with it.

The conversion of photo-to-electric in a DSSC is crucially dependent on available chemical bonds between the dye molecules and nanopowder of TiO_2. Electrons can transfer from excited dye molecules to the conduction band of TiO_2 film through these bonds.

Dye extracted from purple carrot includes one of the anthocyanin compounds. Anthocyanin is a class of pigments found in many plants and is responsible for the colour. There appears red, blue or purple colour depending on the anthocyanin pH. One of the most important advantages of anthocyanin is the binding of carbonyl and hydroxyl groups to the surface of nanopowder layer as shown in Fig. 3. This binding helps in electron transport from the excited anthocyanin molecules to the TiO_2 nanopowder.

The extract of anthocyanin has been investigated in the fabrication of DSSC (Calogero *et al*., 2012). There are many types of anthocyanins from different plants and they give different levels of sensitizing performance (Konczak and Zhang, 2004; Cherepy *et al*., 1997).

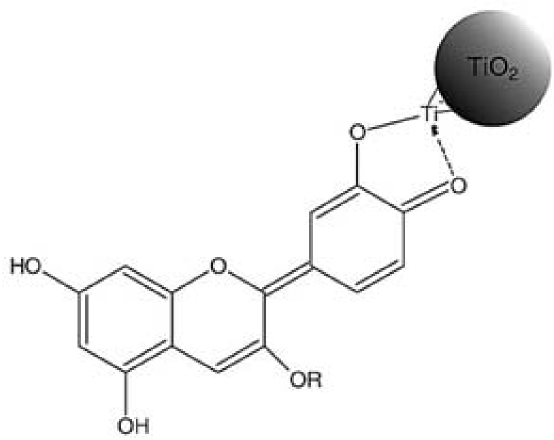

Fig. 3: The basic molecular structure of anthocyanin and its binding with TiO_2 particles.

Kubelka-Munk relationship can be used to study the absorption spectrum shift due to its absorption onto TiO_2 film. Diffuse reflectance spectra were collected with a V-670, JASCO spectrophotometer. The collected data were transformed to absorption spectra according to the Kubelka-Munk relationship. Fig. 4 shows the absorption spectra of the dye extracted from purple carrot root and that of the TiO_2 electrode after being soaked in the solution. The absorption spectrum for the purple carrot dye in ethanol solution shows a peak at 552 nm. As can be seen from the figure, the absorption band of anthocyanin/TiO_2 is red shifted towards higher wavelength (584 nm) compared to that of the anthocyanin in ethanol solution that may be due to complexation between anthocyanin and metal ions, Ti^{4+} (Calogero and Marco, 2008).

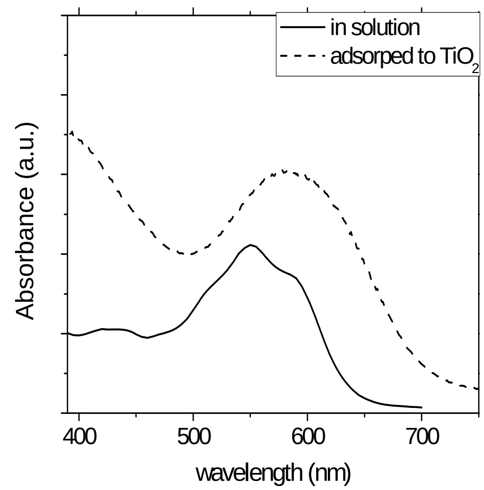

Fig. 4: Absorption spectra of purple carrot extract in ethanol solution and when adsorbed onto TiO_2 film.

Optimization of the extracting temperature of purple carrot dy: The absorption spectra of the extract of purple carrot at different extracting temperatures is shown in Fig. 5. As can be seen from the figure, the absorption peak gets wider by increasing the temperature from 298.15 °K to 323.15 °K. Widening the band of the absorption spectrum results in an increase of photon harvesting, which, in turn, increases the short circuit current.

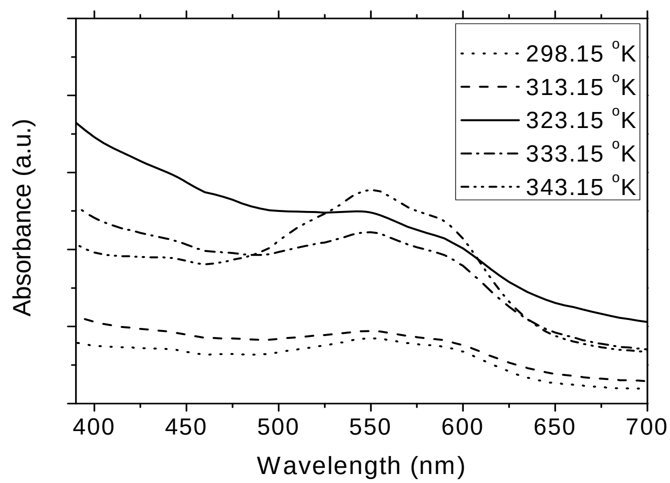

Fig. 5: UV–Vis absorption spectra of the extract of purple carrot at different extracting temperatures.

The J-V of DSSCs fabricated at different extracting temperatures is shown in Fig. 6. Table 2 illustrates the photovoltaic parameters of these cells. The cell efficiency versus the temperature of extraction is shown in Fig. 7, which shows that the efficiency is improved by increasing the extracting temperature from 298.15 °K to 323.15 °K. However, by further increase in extraction temperature, the efficiency decreases. The optimum extraction temperature can be considered 323.15 °K. The increase of the conversion efficiency may be attributed to an increase of the optical absorption of the dye in the range 400 nm - 550 nm at this temperature, as shown in Fig. 6.

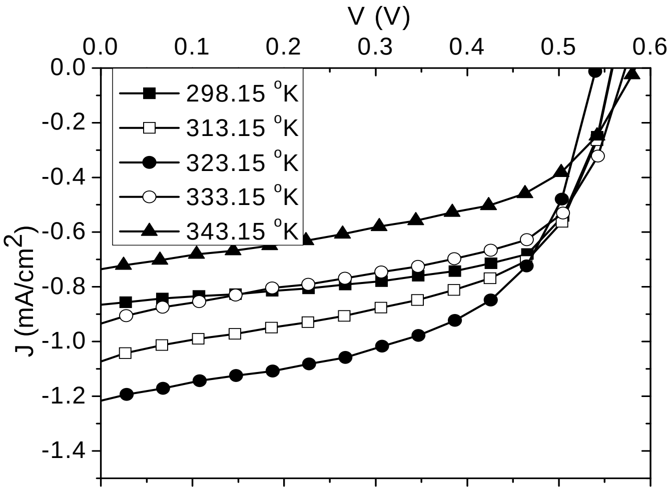

Fig. 6: J-V characteristic curves for DSSCs sensitized at different extracting temperatures.

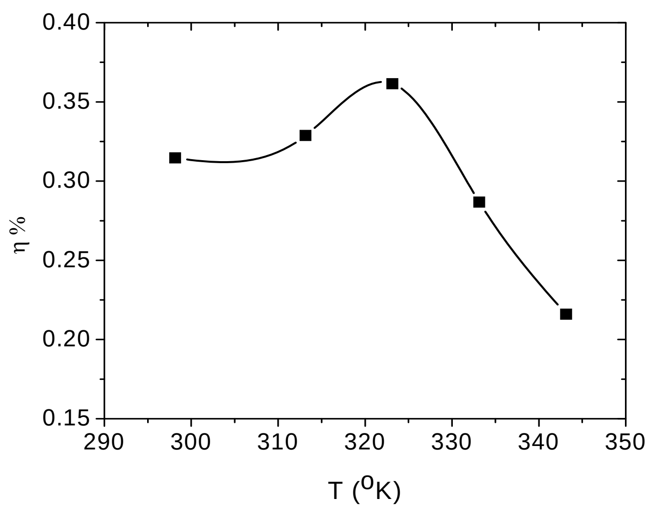

Fig. 7: DSSC efficiency (η) versus the extracting temperature (T) of purple carrot extract.

Table 2: Photovoltaic parameters of the DSSCs sensitized with purple carrot extracted at different temperatures.

Extracting temp. (°K)	J_{sc} (mA/cm^2)	V_{oc} (V)	J_m (mA/cm^2)	V_m (V)	FF	η %
298.15	0.87	0.56	0.68	0.47	0.65	0.31
313.15	1.07	0.56	0.74	0.44	0.55	0.33
323.15	1.21	0.54	0.87	0.42	0.55	0.36
333.15	0.93	0.57	0.62	0.46	0.54	0.29
343.15	0.74	0.59	0.49	0.44	0.50	0.22

Effect of pH of the dye solution: The absorption spectra of purple carrot dye dissolved in ethanol at different pH values, using acetic and hydrochloric acids, are illustrated in Fig. 8. The values of pH of the dye solution using acetic acid were 7.4, 5.8, 4.8, 3.8, and 2.8, whereas, the pH values, using hydrochloric acid, were 7.4, 6.4, 5.0, 3.6, 2.1, and 1.0.

Figure 9 shows the current density-voltage of the DSSCs at various values of pH using acetic and hydrochloric acids. Table 3 shows the most significant parameters of DSSCs at various pH values, using acetic acid.

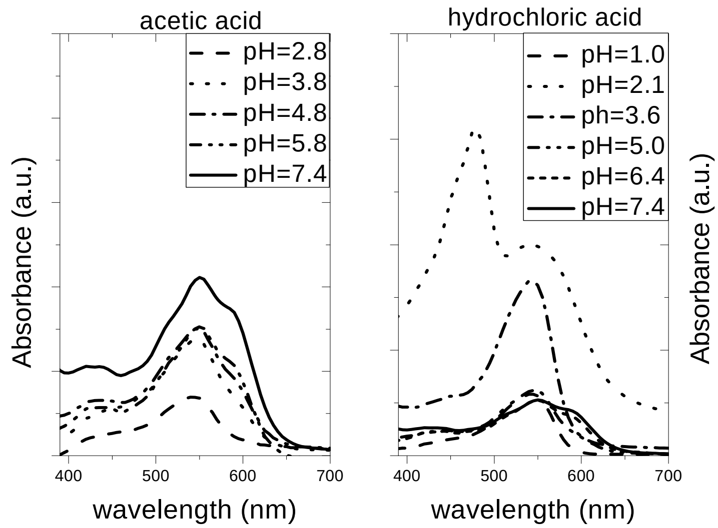

Fig. 8: Absorption spectra of purple carrot extract solutions at various pH values using acetic and hydrochloric acids.

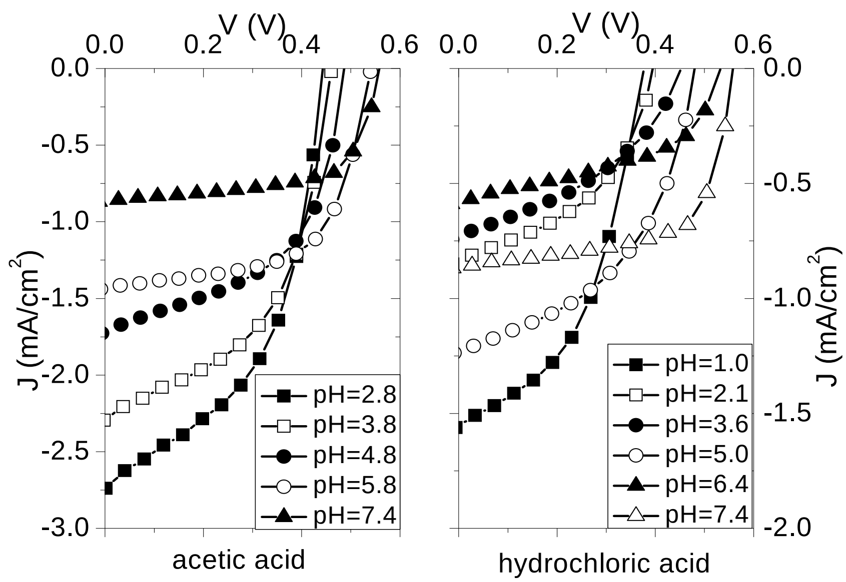

Fig. 9: J-V characteristic curves for DSSCs sensitized with purple carrot extract solutions at various pH values using acetic acid and hydrochloric acid.

Table 3: Photovoltaic parameters of DSSCs sensitized with purple carrot at different pH vales using acetic acid.

pH (Acetic)	J_{sc} (mA/cm^2)	V_{oc} (V)	J_m (mA/cm^2)	V_m (V)	FF	η %
7.4	0.87	0.56	0.68	0.47	0.65	0.31
5.8	1.72	0.49	1.16	0.38	0.52	0.44
4.8	1.44	0.54	1.12	0.43	0.61	0.48
3.8	2.29	0.46	1.59	0.33	0.50	0.53
2.8	2.75	0.44	1.90	0.31	0.49	0.60

The solar cell efficiency versus pH values of the extract solutions, using acetic and hydrochloric acids, are shown in Fig. 10. The efficiency of the solar cell sensitized with purple carrot treated with acetic acid is significantly improved with a decrease of pH of the dye solution from 7.4 to 2.8. The conversion efficiency of the solar cell sensitized with purple carrot treated with hydrochloric acid decreases with decreasing pH from 7.4 to 6.4, while a further decrease of pH from 6.4 to 5, it is enhanced and with a continued decrease of pH from 5 to 3, it decreases. The conversion efficiency exhibits oscillatory behavior with the pH values. The highest conversion efficiency corresponds to pH = 7.4, which is the original value without adding hydrochloric acid. Table 4 shows the most significant parameters of DSSCs fabricated at different pH values, using hydrochloric acid.

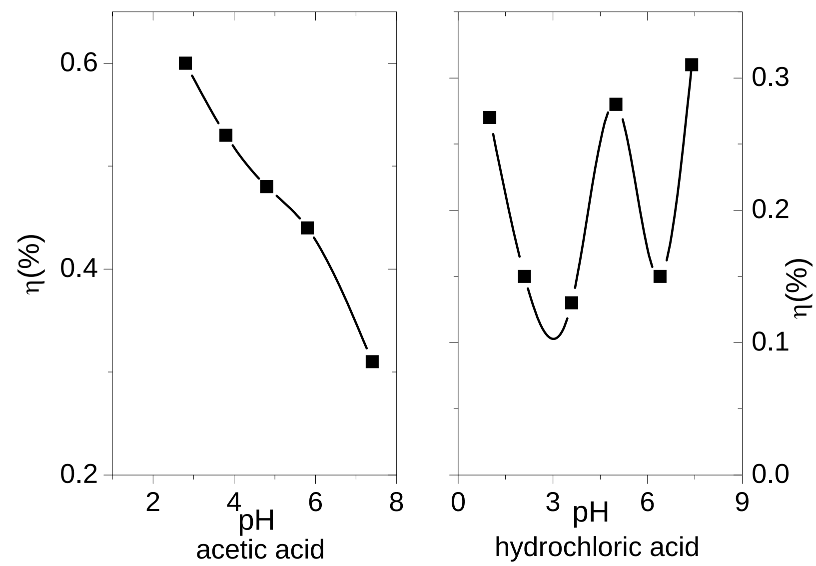

Fig. 10: DSSC efficiency versus the pH of the extract solution of purple carrot using acetic and hydrochloric acid.

Table 4: Photovoltaic parameters of DSSCs sensitized by purple carrot at different pH vales, using hydrochloric acid.

pH (Hydrochloric)	J_{sc} (mA/cm^2)	V_{oc} (V)	J_m (mA/cm^2)	V_m (V)	FF	η %
7.4	0.87	0.56	0.68	0.47	0.65	0.31
6.4	0.58	0.38	0.36	0.40	0.67	0.15
5.0	1.23	0.48	0.84	0.33	0.47	0.28
3.6	0.73	0.45	0.44	0.30	0.40	0.13
2.1	0.84	0.40	0.57	0.27	0.46	0.15
1.0	1.55	0.38	1.08	0.25	0.46	0.27

The acetic acid treatment of the dye solutions to adjust pH values improved the cell conversion efficiency whereas, treating the cells with hydrochloric acid to adjust pH, resulted in lower conversion efficiency because hydrochloric acid is a strong acid as compared to acetic acid. When treating DSSCs with acetic acid, the absorption spectrum exhibits no shift but it modifies the TiO_2 nanopowder surface. These results agree with Hao *et al*. (2004), where the treatment with acetic acid showed improved efficiency than the untreated ones. The acetic acid modified TiO_2 layers have a better morphology, which results in an increase in the short circuit current and better transportation of electrons (Hao *et al*., 2004). The treatment with hydrochloric acid showed relatively poor performance. The presence of hydrochloric acid in the dye changes the surface state of the TiO_2 and creates defect centers on its surface. This effect prevents the transport of electrons from the excited level of the dye to the TiO_2 grid (Jeong *et al*., 2010).

Electrochemical impedance spectroscopy: Electrochemical impedance spectroscopy was carried out for the DSSCs at pH 2.8. The pH was adjusted using acetic acid. The electrochemical impedance spectroscopy was conducted using AUT 85276 Potentiostat-Galvanostat with frequency response analyzer FRA 32 Module device. Fig. 11 shows Nyquist plots for these DSSCs under an illumination of 100 mW/cm^2 at –0.4 V, –0.6 V, and –0.8 V applied voltages.

The electrochemical semicircle fit was obtained for these DSSCs as illustrated in Fig. 12. An equivalent circuit has been obtained for the DSSCs and its components are presented in Table 5.

According to Table 5, there is an increase in the charge-transfer resistance (R_{CT}) when the cell is illuminated at –0.4 V applied voltage, however, no difference is observed at –0.6 V and –0.8 V in the R_{CT} when the cell is illuminated (Table 5).

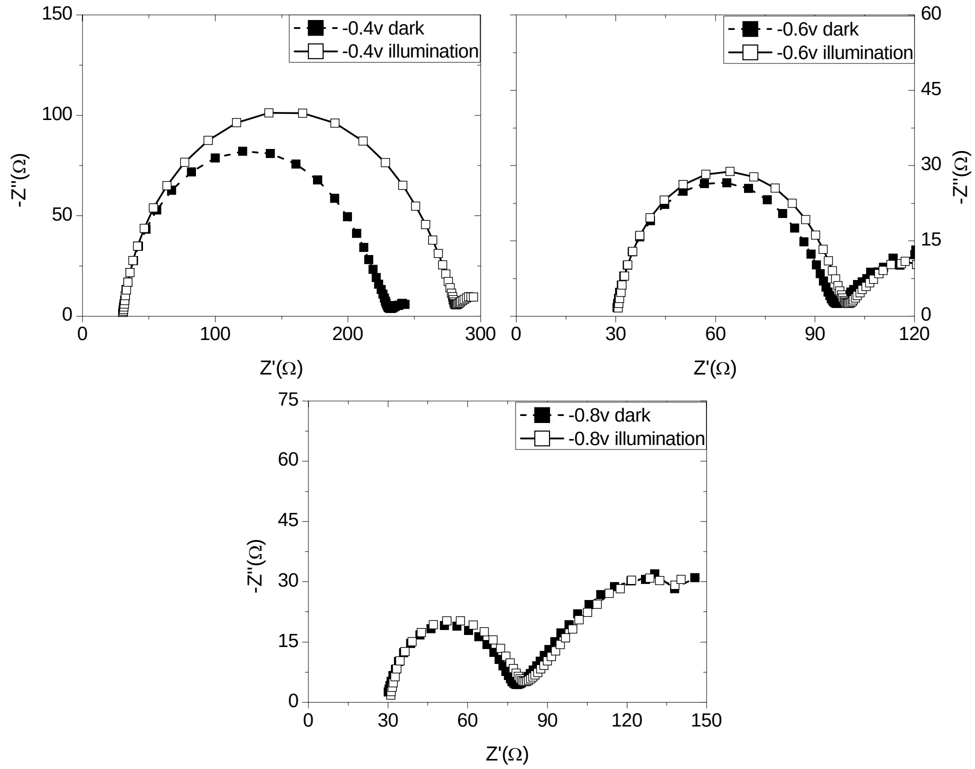

Fig. 11: Nyquist plots of DSSC sensitized by purple carrot of pH 2.8 with acetic acid at –0.4 V, –0.6 V, and –0.8 V in the dark and under an illumination of 100 mW/cm^2.

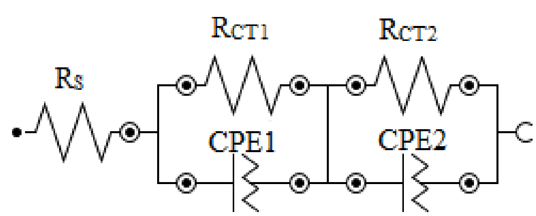

Fig. 12: The equivalent circuit for DSSC sensitized by purple carrot of pH 2.8 with acetic acid.

Table 5: EIS results from data-fitting of Nyquist plots to the equivalent circuit model for the DSSC sensitized by purple carrot of pH 2.8 with acetic acid.

pH 2.8 with acetic acid	R_S (Ω)	R_{CT1} (Ω)	C_1 (μF)	R_{CT2} (Ω)	C_2 (μF)
–0.4v dark	30.1	28.10	149.87	191	2.50
–0.4v illumination	30.1	245	1.18	50.9	45625.82
–0.6v dark	31.7	62.7	0.93	24.3	11900
–0.6v illumination	31.9	66.1	0.92	23.7	14300
–0.8v dark	29.4	46.9	0.86	141	8966.09
–0.8v illumination	30.5	47.1	0.87	183	13585.55

Fig. 13 shows the Bode plot, which is another representation of the impedance that provides directly the electron lifetime for the processes in the DSSCs and Table 6 presents the electron lifetime for the these processes. The reduction obtained in the value of R_{CT} means that there is a decrease in the recombination rate and it indicates fast electron-transfer processes in the DSSCs at pH 2.8 adjusted with acetic acid compared with the untreated cell. The efficient charge-transfer paths decrease the recombination rate of electrons with I_3^- or the oxidizing dye, resulting in a high photocurrent density and conversion efficiency

(Zhong *et al*., 2011). The decrease in the life time may be attributed to easier electron transfer within the photoelectrodes and subsequent charge transfer at photoelectrode/FTO interface, which is beneficial for both electron generation and transport (Table 6).

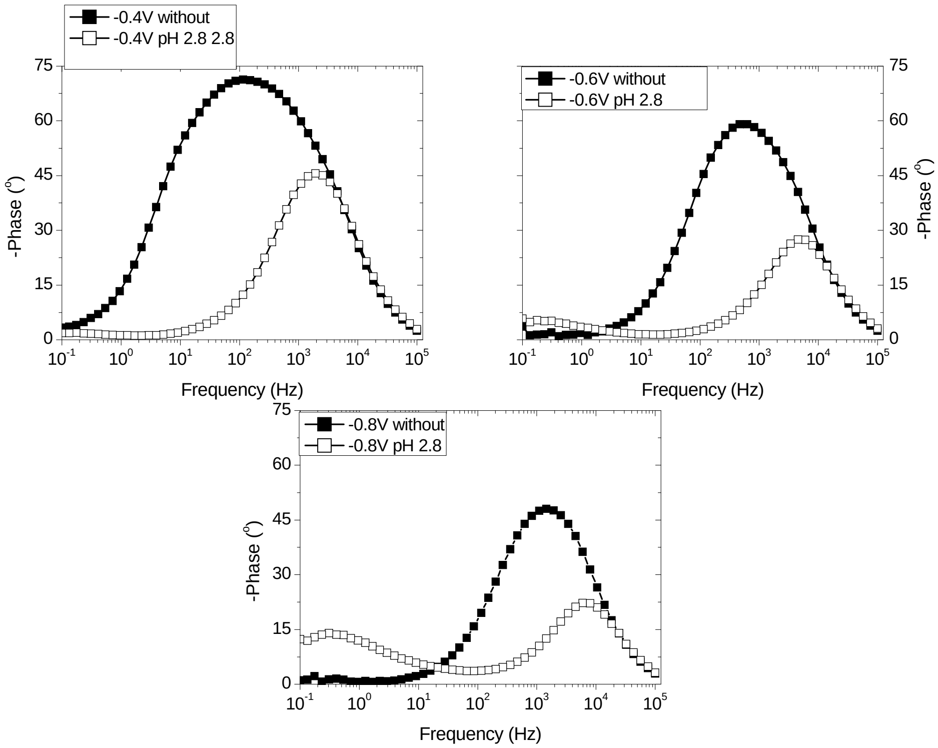

Fig. 13: Bode plots of DSSCs sensitized by purple carrot without treatment and DSSC sensitized by purple carrot of pH 2.8 with acetic acid under an illumination of 100 mW/cm^2 at –0.4V, –0.6V and –0.8V applied voltages.

Table 6: Electron lifetime calculations from Bode plots.

Treatment	f (frequency corresponding to peak in Bode plot) (Hz)	τelectron (ms)
–0.4V without	115.40	1.38
–0.4V pH 2.8	1930.70	0.08
–0.6V without	625.06	0.25
–0.6V pH 2.8	4498.40	0.04
–0.8V without	1456.30	0.11
–0.8V pH 2.8	5963.60	0.03

CONCLUSION

Natural dyes extracted from the roots of purple carrot, beet, and curcuma, were used as sensitizers for DSSCs. The DSSCs based on purple carrot have shown the best performance. The optimum temperature for extracting the dye was investigated and found to be 323.15 °K. The cell performance improved, by adjusting the dye pH with acetic acid. The efficiency improved significantly by decreasing pH from 7.4 to 2.8 which corresponds the highest efficiency. The impedance spectroscopy confirms the enhancement of the efficiency, due to better charge transport and injection within the photoanode.

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