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Dyes Extracted From the Seeds of Eruca Sativa, Nigella Sativa and Ammi Visnaga as Photosensitizers for Dye-Sensitized Solar Cells

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Abstract: In this work, the extracts of three natural dyes obtained from the seeds of Eruca Sativa, Nigella sativa, and Ammi visnaga were used as photosensitizers for dye-sensitized solar cells (DSSCs). Dye Extraction was carried out before and after grinding the plant seeds. TiO_2 nanopowder was used as a semiconducting material. The UV-Vis absorption spectra of the extracts of the seeds of Eruca Sativa, Nigella sativa, and Ammi visnaga in ethyl alcohol solution and the current-voltage characteristic curves of the fabricated DSSCs were measured. The energy gaps of the extracted dyes were estimated. The best response was obtained for the DSSC sensitized with Ammi visnaga with an efficiency of 0.107% before grinding the raw material and 0.132% after grinding. The acidic treatment of FTO glass sheets with HNO_3 improved the efficiency by 123%, 108% and 105% for the three extracts of Eruca Sativa, Nigella sativa and Ammi visnaga, respectively.

Keywords: dye-sensitized solar cells, plant seeds, acidic treatment, impedance spectroscopy.

1 Introduction

Dye-sensitized solar cells (DSSCs) have been widely studied because of their simple fabrication processes compared with conventional silicon solar cell and lower production cost. The photoelectric conversion efficiency of DSSCs can reach up to 10%, whereas that of silicon solar cell is about 20% [1]. DSSCs are considered one of the recent types of photovoltaic devices for the conversion of sunlight into electrical energy [2]. This type of solar cells lies between solid state cell and a regenerative photoelectrochemical cell. In DSSC device, the dye plays a key role as a sensitizer which has the ability to absorb the sunlight and produce photo-excited electrons at the interface of the semiconductor material. The DSSCs have been fabricated with different semiconductor oxide materials, such as TiO_2, Nb_2O_5, ZnO and SnO_2 [3-6]. Among them, the TiO_2 oxide is the most popular because it is inexpensive material with very high dielectric constant and has high refractive index. Moreover, it is nontoxic, readily available, and stable photo-electrode in photo-electrochemical cells [7-9]. The DSSCs efficiency and stability depend on different parameters such as the efficiency of dye absorption, band gap, electron transport, surface area, and the porosity of the semiconductor materials [10-12].The TiO_2 nanoparticles offer most of these parameters such as the porous structure, the high surface area and appropriate band gap (3.2 eV). Natural dyes are of interest because of many features such as easy preparation, inexpensive nature, nontoxicity, environmentally friendly, complete biodegradability and wide availability. Several previous studies investigated natural dyes such as pomegranate juice [9], black grapes [13], red amaranth [14], red turnip [15], hibiscus [16], chlorophyll [17,18], dried plant leaves [19-21], plant seeds [22], and others [23] as sensitizers in dye-sensitized solar cells. Adriano et al. [24] used nature dyes obtained from java plum (syzigium cumin), red cabbage (brassica oleracea), hibiscus rosa-sinensis flower, and begonia red leaves as sensitizers of DSSCs. The photoelectrochemical performance of the DSSCs based on these dyes showed conversion efficiencies ranged from 0.50% to 0.94%. Reena et al. [25] investigated four natural dyes: the leaves of teak (tectona grandis), tamarind (tamarindus indica), eucalyptus (eucalyptus globulus), and the flower of crimson bottle brush (callistemon citrinus). The leaves of teak have shown the best performance. Jasim et al. [14] investigated the effects of the solvents polarities of normal water, boiling water, ethanol, methanol, and acetone in the dye extraction. They found the acetone performed a good solvent for the dye extraction.

In this paper, DSSCs were fabricated using TiO_2 nanoparticles as a semiconducting material and natural dyes obtained from the seeds of Eruca Sativa, Nigella sativa, and Ammi visnaga. Thin films of TiO_2 were spread on transparent conducting FTO coated glass using doctor blade

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method. The extracted dyes in ethyl alcohol solution were characterized by UV–Vis absorption spectra. The current density-voltage (J-V) characteristic curves of the cells were measured and the efficiency of each DSSC related to each dye was calculated. The natural dyes extracted from the raw materials before and after grinding were used to fabricate the DSSCs. Moreover, the effect of acidic treatment of FTO sheets on the photovoltaic properties was reported.

2 Experimental

2.1 Extraction of natural dye sensitizers

The natural dyes used in this study were extracted from the following plant seeds: Eruca Sativa, Nigella sativa, and Ammi visnaga. The raw natural materials were first washed with distilled water several times to free from dust, and then dried at 70 °C. The process of dye extraction was carried out before and after grinding the dyes so that two extracts were obtained for each dye: one was obtained before grinding and the other after grinding the raw material. 1 gm from each material (before grinding) was immersed in 5 ml ethyl alcohol at room temperature and in dark for one day. Other amounts of the dried materials were grinded into fine powder in a mortar. 1 gm from each material powder was immersed in 5 ml ethyl alcohol at room temperature and in dark for one day. After filtration of the solutions of natural dyes were obtained. Dye solutions were protected from direct light exposure.

2.2 Preparation of dye sensitized solar cells

FTO conductive glass with sheets of resistance of 15 Ω/cm^2 and transmission >80% (Xinyan Technology Ltd., Hong Kong) were cut into rectangle pieces of dimensions 0.8 cm×1.6 cm with a glass cutter. The samples were cleaned ultrasonically using acetone, ethanol and distilled water, and then dried in an oven at 60 °C for 30 min. If the cleaned FTO-glass was not directly used for cell assembly, it was stored in ethanol. The TiO_2 paste was prepared by grinding 0.05 gram of nanocrystalline TiO_2 powder (P25 titanium oxide; Evonik Degussa Japan Co., Ltd., Tokyo, Japan) and 0.1g of polyethylene glycol in a mortar for half an hour until a homogeneous paste was obtained [26].

This paste was coated on FTO sheets by doctor blade method to form a thin layer with an effective area of 0.5cm×0.5cm. These films were then dried in an oven at 70 °C for 20 min and sintered in another oven at 450 °C for 40 min then cooled down to 70 °C. The TiO_2 sintered films were immersed in the dye solution and kept at room temperature (≈ 25°C) for one day under dark and then dried. The dye-absorbed TiO_2 electrode was sandwiched together with a platinum -coated FTO glass counter electrode. Finally, the liquid electrolyte containing iodide/tri-iodide (I^-/I^3) redox was injected to fill the space between the cell electrodes.Another group of FTO conductive glass sheets were immersed in one of three different acidic solutions of HNO_3, H_2SO_4 and H_3PO_4, each of 0.1 M concentration for 10 min. The TiO_2 paste was coated on the acid treated FTO samples and another set of DSSCs was prepared as described above.

2.3 Characterization and measurement

The UV–Vis absorption spectra of the extracts of Eruca Sativa, Nigella sativa, and Ammi visnaga in ethyl alcohol solution were measured using Thermoline Genesys 6 UV-Vis spectrophotometer. The wavelength range of absorption spectra analysis was taken from 400−850 nm. The current density-voltage (J-V) characteristic curves of the fabricated DSSCs were measured using National Instruments data acquisition card (USB NI 6251) in combination with a Labview program. The J-V curves were measured at 100 mW/cm^2 irradiations using high pressure mercury arc lamp. The voltage step and delay time for the measurement were 10 mV and 40 ms, respectively. All measurements were performed at room temperature.

3 Results and discussion

3.1 Absorption of natural dyes

The UV-Vis absorption spectra of Eruca Sativa, Nigella sativa and Ammi visnaga extracts are shown in Fig. 1. In general the three dyes have obvious analogous absorption peaks in the blue green visible region. Eruca Sativa has a peak at 521.1 nm. Nigella sativa shows an absorption peak at 507 nm and the peak of Ammi visnaga was observed at 538 nm. These results were employed for the calculation of the energy gaps for the natural extracts as shown in Fig. 2 and using the well-known equation $E = \frac{hc}{\lambda}$, where E = absorbed photon energy, h = 6.63 x 10^{-34} J.s, c =3.0 x 10^8 m/s, and λ is the wavelength. So it is clear that Eruca Sativa, Nigella sativa and Ammi visnaga have the energy gaps of 2.38 eV, 2.45 eV and 2.31 eV, respectively.

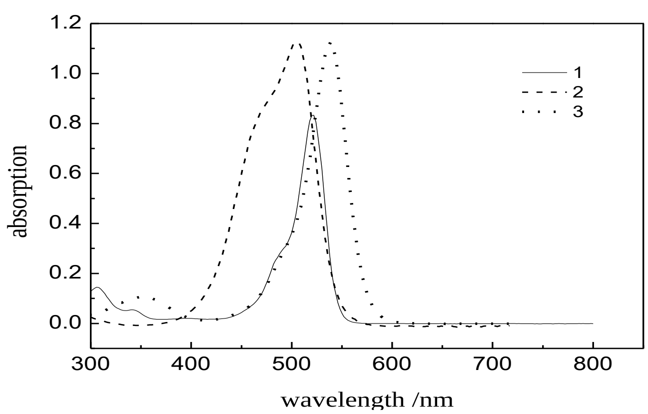

Figure 1: UV-Vis absorption spectra of (1) Eruca Sativa, (2) Nigella sativa, and (3) Ammi visnaga in ethyl alcohol solution.

Table 1: Photoelectrochemical parameters of the DSSCs sensitized by the three extracts of Eruca Sativa, Nigella sativa and Ammi visnaga, before and after grinding.

Natural dye	J_{sc} (mA/cm^2)		V_{oc} (V)		FF(%)		η(%)	
	before	after	before	after	before	after	before	After
Eruca Sativa	0.239	0.287	0.497	0.486	59.3	63.6	0.071	0.088
Nigella sativa	0.200	0.241	0.405	0.447	58.6	59.7	0.047	0.064
Ammi visnaga	0.321	0.332	0.529	0.610	63.0	65.2	0.107	0.132

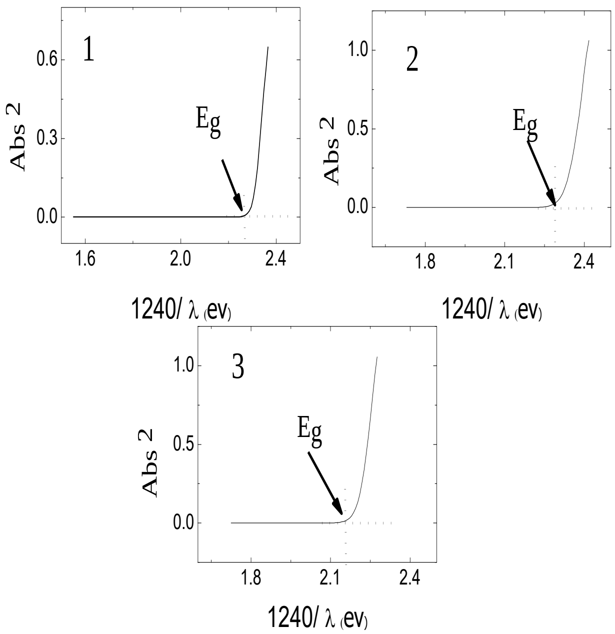

Figure 2: The optical energy gap of (1) Eruca Sativa, (2) Nigella sativa, and (3) Ammi visnaga.

3.2 Photovoltaic properties

The measurements of current density and voltage of DSSCs fabricated using the natural sensitizers of Eruca Sativa, Nigella sativa and Ammi visnaga seeds before and after grinding was carried out under 100 mW/cm^2 illumination. From the J-V characteristic curves illustrated in Fig. 3, the photoelectrochemical parameters of the solar cells under study were presented in Table 1. These parameters consist of short circuit current J_{sc}, open circuit voltage V_{oc}, fill-factor FF, and conversion efficiency η. It is clear from the listed results in Table 1 that the DSSCs sensitized with grinded materials exhibit better performance in all parameters with almost similar improvement percentage. Moreover the photovoltaic response of the cells anchored with Ammi visnaga was the best in which J_{sc} = 0.321 mA/cm^2, V_{oc} = 0.529 V, FF = 63%, and η = 0.107% before grinding the raw material and J_{sc} = 0.332 mA/cm^2, V_{oc} = 0.610 V, FF = 65.2%, and η = 0.132% after grinding with efficiency improvement of about 123% for the grinded material.

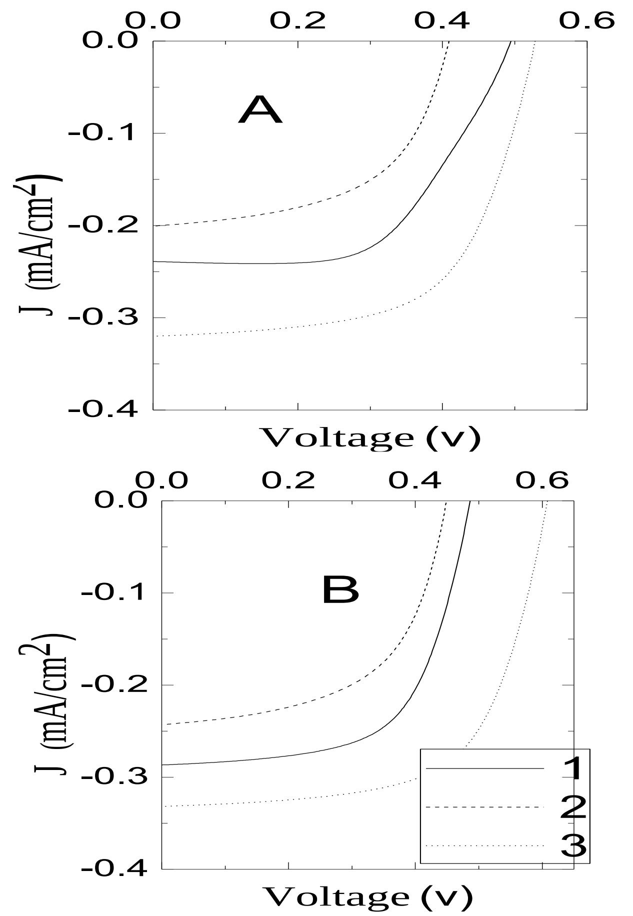

Figure 3: Current density–voltage curves for the DSSCs sensitized by (1) Eruca Sativa, (2) Nigella sativa and (3) Ammi visnaga. (A) before grinding and (B) after grinding.

3.3. Photovoltaic properties after FTO acidic treatment

The current density-voltage characteristic curves of the acidic pretreated FTO glass DSSCs and sensitized by the three natural dyes extracted from Eruca Sativa, Nigella sativa and Ammi visnaga are shown in Fig. 4. Table 2 gives a full summary of their photoelectrochemical parameters. The data presented in Table 2 shows a better performance after the acidic treatments in general. DSSCs sensitized with Eruca sativa exhibit efficiency improvements of 123%, 114% and 105% with HNO_3, H_2SO_4 and H_3PO_4 acids, respectively.

On the other hand, DSSCs sensitized with Nigella sativa has an efficiency improvement of 108% with HNO_3 and H_2SO_4 treatments. Ammi visnaga recorded η □=0.139, J_{sc} = 0.402 mA/cm^2, V_{oc} = 0.618 V with HNO_3, which means it is still has the highest performance before and after acidic treatment with 105% efficiency rise, and no change with H_2SO_4 and H_3PO_4. So we can say that the treatment of FTO with HNO_3 progressed the efficiency by 123%, 108% and 105% for the three extracts Eruca Sativa, Nigella sativa and Ammi visnaga, respectively.

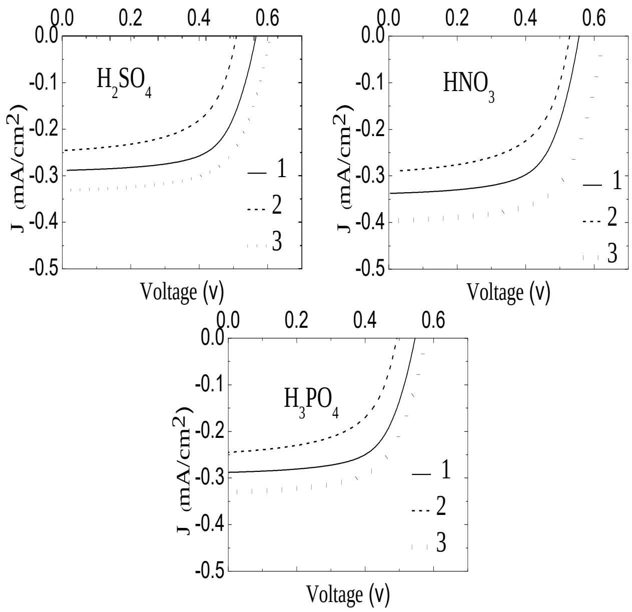

Figure 4: Current density–voltage curves for the DSSCs sensitized by (1) Eruca Sativa, (2) Nigella sativa, (3) Ammi visnaga using acid treated DSSCs

Table 2: Photoelectrochemical parameters of the acidic treated DSSCs sensitized by the three extracts of Eruca Sativa, Nigella sativa and Ammi visnaga.

Natural dye	Acids	J_{sc}(mA/cm^2)	V_{oc} (volt)	η %
Eruca Sativa	H_2SO_4	0.293	0.575	0.101
	HNO_3	0.355	0.582	0.109
	H_3PO_4	0.294	0.577	0.093
Nigella sativa	H_2SO_4	0.297	0.523	0.069
	HNO_3	0.297	0.523	0.069
	H_3PO_4	0.250	0.507	0.060
Ammi visnaga	H_2SO_4	0.357	0.608	0.131
	HNO_3	0.402	0.618	0.139
	H_3PO_4	0.332	0.585	0.132

3.4. Impedance spectroscopy measurements

Electrochemical impedance spectroscopy (EIS) study was employed to understand the main features of the charge transport kinetics in the cell. The EIS study was obtained by applying a small AC voltage signal of amplitude 10 mV with varying frequency (10 mHz – 10 MHz) to the DSSCs under study. Figure 5 represents the Nyquist curves of the solar cells sensitized by Eruca Sativa, Nigella sativa and Ammi visnaga whereas Fig. 6 illustrates Bode modulus curves for the DSSCs sensitized by the three extracts. According to the Nyquist and Bode curves, the series resistance R_S, the charge transfer resistance according to recombination R_{CT}, a constant phase element CPE due to double layer capacitance (C_{dl}), the effective lifetime of electrons τ, and the CPE exponential coefficient α can be determined. All these parameters are listed in Table 3. From the results presented in Table 3, it is clear that R_S is much smaller than R_{CT} for all the tested DSSCs dyed with Eruca Sativa, Nigella sativa and Ammi visnaga which means fast electron transport and difficulty for charge recombination process leading to long lifetime (within millisecond range) for electrons in the film. This leads to a higher DSSC efficiency. The CPE exponential coefficient α values for all the cells are almost equal (0.6-0.65) and gives a sign of diffusion with deviations and surface roughness.

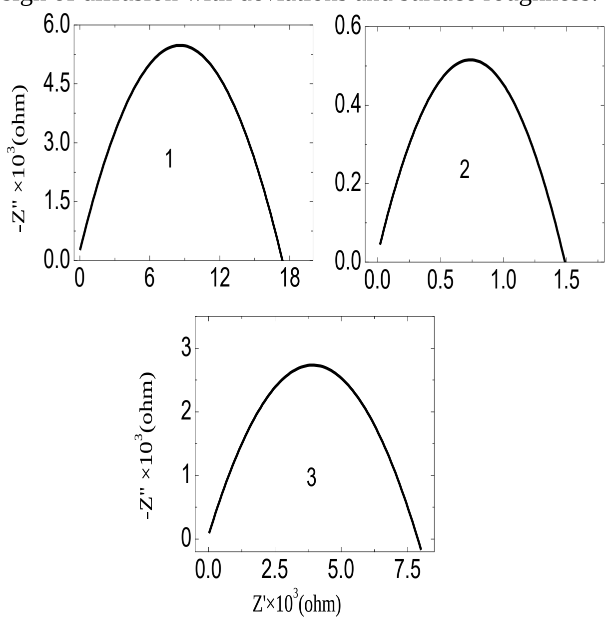

Figure 5: EIS Nyquist plots of DSSCs sensitized by (1) Eruca Sativa, (2) Nigella sativa, and (3) Ammi visnaga.

Table 3: The impedance spectroscopy parameters of the DSSCs sensitized by the extracts of Eruca Sativa, Nigella sativa and Ammi visnaga.

Natural dye	**R_S (KΩ)**	**R_{CT} (KΩ)**	**C_{dl} (μF)**	**α (CPEcoefficient)**	**τ (ms)**
Eruca Sativa	0.025	17.41	0.876	0.64	15.2
Nigella sativa	0.017	1.495	7.702	0.65	11.5
Ammi visnaga	0.052	8.018	0.331	0.60	2.55

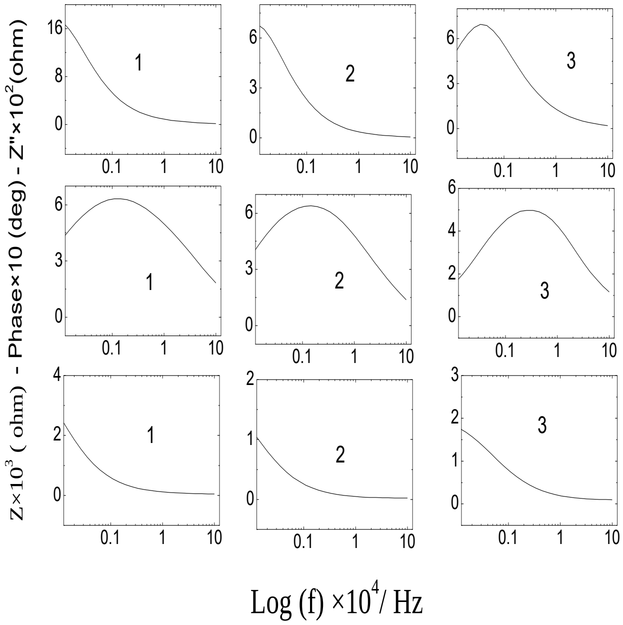

Figure 6: Bode plots of DSSCs sensitized by (1) Eruca Sativa, (2) Nigella sativa, and (3) Ammi visnaga.

4 Conclusions

Dye-sensitized solar cells (DSSCs) were fabricated using TiO_2 nanoparticles and natural extracts of Eruca Sativa, Nigella sativa, and Ammi visnaga seeds. The UV-Vis absorption spectra of the three extracts showed that they have obvious analogous absorption peaks in the blue green visible region. The absorption spectra results were employed to calculate the energy gaps of the natural extracts and it was found that Eruca Sativa, Nigella sativa and Ammi visnaga have the energy gaps of 2.38 eV, 2.45 eV and 2.31 eV, respectively. The J-V characteristic curves of the fabricated DSSCs were carried out and the photoelectrochemical parameters of the solar cells were calculated. The efficiencies of the DSSCs sensitized with the extracts obtained before grinding the seeds were 0.071, 0.047, and 0.107 for Eruca Sativa, Nigella sativa, and Ammi visnaga seeds, respectively. These efficiencies reached 0.088, 0.064 and 0.132 after grinding the raw seeds. The FTO glass sheets were then treated using one of three acids. The treatment with HNO_3 led to an efficiency enhancement of 123%, 108% and 105% for the three extracts of Eruca Sativa, Nigella sativa and Ammi visnaga, respectively. Electrochemical impedance of the fabricated DSSCs was carried out and Nyquist and Bode curves were plotted. The EIS parameters were calculated from which we may conclude there is fast electron transport and difficulty for charge recombination process leading to long lifetime for electrons in the film.

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