Aldimine derivatives as photosensitizers for dye-sensitized solar cells

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Abstract: Five aldimine derivatives were prepared by condensation of the appropriate amine with salicylaldehyde (m1–m4) and 4-aminobenzoic acid with 2-thiophene carboxaldehyde (m5). A molar ratio of 1:1 was used and the mixture was refluxed in ethanol for 2 h to obtain the corresponding aldimine derivative. These derivatives were used as photosensitizers for dye-sensitized solar cells (DSSCs). The best performance was obtained for the DSSC sensitized with 2-(2-hydroxybenzylideneamino)benzoic acid, for which a short-circuit current of 2.86 mA/cm^2, an open-circuit voltage of 0.562 V, and an efficiency of 0.575% were obtained.

Key words: Dye-sensitized solar cell, TiO$_2$, aldimine derivatives

1. Introduction

The direct conversion of sunlight into electrical power by solar cells is one of the most promising future energy production techniques. Different from many other electrical energy production techniques, solar cells are of particular interest because electricity is produced without the exhaust of greenhouse gases. Dye-sensitized solar cells (DSSCs) were first announced as an innovative solar energy conversion technology in 1991. They are found to have many advantages, such as that they are a cheap, renewable, and environmentally acceptable source of energy production [1–5]. A DSSC has a very simple structure. It consists of a sensitizing dye chemically anchored to a nanocrystalline wide band-gap semiconducting material, such as TiO$_2$ or ZnO. A high surface area for dye coverage is provided by the mesoporosity of the semiconducting material. This allows the capturing of the majority of the incident solar energy by the adsorbed dye monolayer. The cell is illuminated through a transparent conductive oxide electrode such as fluoride-doped tin oxide (FTO). The semiconducting material in paste formulation is deposited onto the FTO layer. The porous photoanode is usually immersed in a redox electrolyte I^-/I_3^- and is electrically connected to a counter electrode. When the DSSC is illuminated, electrons are absorbed by the dye and injected into the semiconductor conduction band, and they move through the FTO substrate to the external load. The oxidized dye is reduced by the electrolyte, which transports the positive charges to the counter electrode.

The efficiency of a DSSC depends on many factors, such as a delicate balance of the kinetics for injection, dye regeneration, and recombination reactions [4]. The sensitizing dye behaves as a solar energy absorber. An ideal sensitizer should have a set of features to make the DSSC efficient. It should absorb all the light below a threshold wavelength of about 920 nm. Moreover, it should be firmly grafted to the semiconducting material

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surface and inject electrons into the conduction band with a quantum yield of unity. Another important parameter is the dye redox potential, which should be high enough so that it can be regenerated rapidly via electron donation from the electrolyte. Finally, it should be stable enough to sustain at least 10^8 redox turnovers under illumination, corresponding to about 20 years of exposure to natural light. The best photovoltaic performance in terms of both conversion yield and long-term stability has so far been achieved with polypyridyl complexes of ruthenium and osmium. The sensitizers used in DSSCs are divided into 2 types, organic and inorganic dyes, according to the structure. Inorganic dyes include metal complexes, such as polypyridyl complexes of ruthenium and osmium, metal porphyrin, phthalocyanine, and inorganic quantum dots, while organic dyes include natural and synthetic organic dyes. So far, devices based on ruthenium polypyridyl sensitizers and an iodide/triiodide redox mediator exhibit an efficiency of about 11% [6]. Many researchers have developed synthetic dyes for the fabrication of efficient DSSCs [7,8]. Dai et al. presented DSSCs fabricated using TiO_2 photosensitized by adsorbed anthocyanin dyes with iodide/iodine in water at pH 1 [7]. In 2009, the use of a diazapentadiene derivative and 2 compounds of triazole derivatives as photosensitizers of DSSCs was achieved [8]. Intensive research has been carried out to fabricate DSSCs using natural dyes [9,10].

In this work, DSSCs are fabricated and studied using aldimine derivatives as sensitizing dyes. These dyes were prepared by condensation of the appropriate amine with salicylaldehyde or thiophene carboxaldehyde. A molar ratio of 1:1 was used and the mixture was refluxed in ethanol for 2 h to obtain the corresponding aldimine derivative.

2. Experiment

2.1. Preparation of dye sensitizers

Five aldimine derivatives (Figure 1) were prepared by condensation of the appropriate amine with salicylaldehyde (m1–m4) and 4-aminobenzoic acid with 2-thiophene carboxaldehyde (m5). A 1:1 molar ratio was used and the mixture was refluxed in ethanol for 2 h to obtain the corresponding aldimine derivative. The obtained derivatives were 2-((phenylimino)methyl)phenol (m1), 2-(2-hydroxybenzylideneamino)benzoic acid (m2), 2-(2-hydroxybenzylideneamino)phenol (m3), 4-(2-hydroxybenzylideneamino)benzoic acid (m4), and N-(thiophen-2-ylmethylene)aniline (m5).

2.2. Preparation of dye-sensitized solar cells

FTO conductive glass sheets with sheet resistance of 15 Ω/square and transmission of >80% (Xinyan Technology Ltd., Hong Kong) were first cut into small pieces of dimensions 1 cm $\times$ 1 cm. The pieces were cleaned in an ultrasonic bath for 15 min, rinsed with water and ethanol, and then dried. TiO_2 paste was prepared by adding 50 mg of TiO_2 nanopowder, 10–25 nm (US Research Nanomaterial, Inc., USA), to 50 mg of polyethylene glycol; the mixture was then ground for 30 min in a mortar until a homogeneous paste was obtained. The paste was deposited on the FTO conductive glass by doctor-blade technique in order to obtain a TiO_2 layer of 0.25 cm^2 in area. The TiO_2 layer was preheated at 60 $^\circ$C for 20 min and then sintered at 450 $^\circ$C for 30 min. After cooling down to 60 $^\circ$C, the sheets with TiO_2 thin film layer were immersed in a solution of the aldimine derivatives for 24 h. The dyed TiO_2 layer and a sputtered platinum counter electrode were assembled to form a solar cell by sandwiching a redox (I^-/I_3^-) electrolyte solution. The electrolyte solution was composed of 2 mL of acetonitrile, 8 mL of propylene carbonate, 0.668 g of KI, and 0.0634 g of I_2.
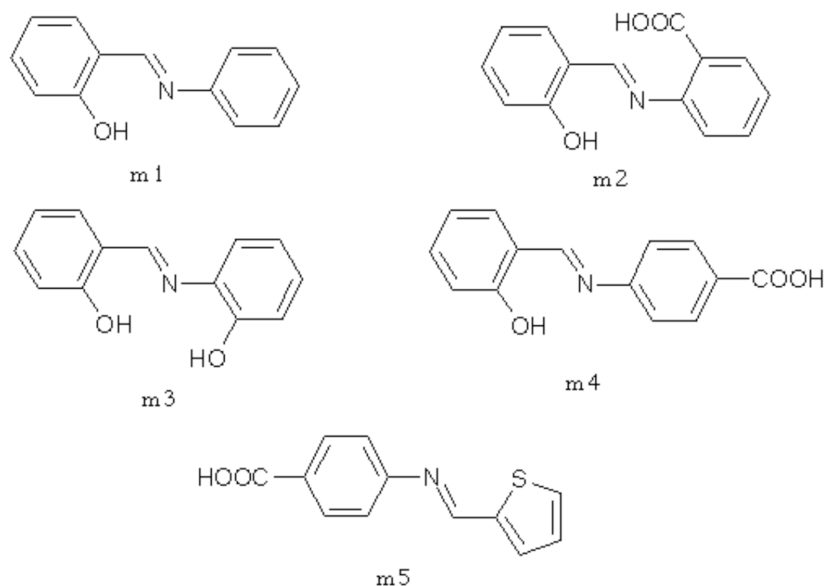
Figure 1. Chemical structures of the aldimine photosensitizers.

2.3. Measurements

The UV-VIS absorption spectra of all aldimine derivatives were measured using a UV-VIS spectrophotometer (GENESYS 6, Thermo Scientific, USA). The absorption spectrum analysis was carried out in the wavelength range from 400 to 800 nm. The $J-V$ characteristic curves under illumination were established using a National Instruments data acquisition card (USB NI 6251) in combination with the LabVIEW program. The $J-V$ curves were measured at 100 mW/cm^2 irradiation using a high-pressure mercury arc lamp.

3. Results and discussion

Figure 2 illustrates the absorption spectra of 2-((phenylimino)methyl)phenol (m1), 2-(2-hydroxybenzylideneamino)benzoic acid (m2), 2-(2hydroxybenzylideneamino) phenol (m3), 4-(2-hydroxybenzylideneamino)benzoic acid (m4), and N-(thiophen-2-ylmethylene)aniline (m5) using ethyl alcohol as a solvent. As can be seen from the figure, the absorption peaks of all dyes lie in the spectral region having a wavelength of less than 500 nm.
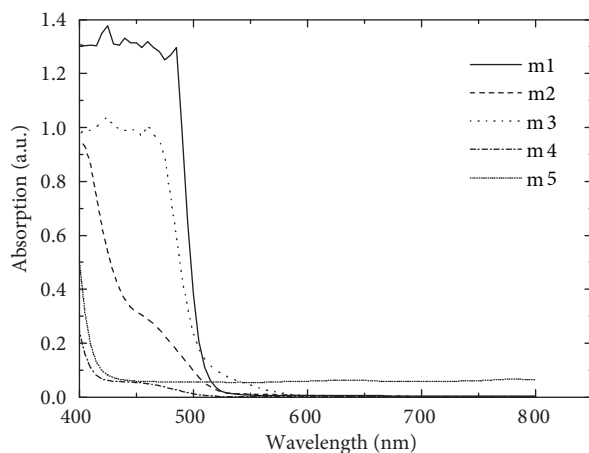

Figure 2. The absorption spectra of all aldimine derivatives.

The J-V characteristic curves of the 5 DSSCs dyed with aldimine derivatives were established under illumination with white light of intensity 100 mW/cm^2 from a high-pressure mercury arc lamp. These curves are shown in Figure 3. The DSSC output power was calculated as $P = JV$ using the $J - V$ data corresponding to each cell. The output power plotted as a function of V is shown in Figure 4 for all cells. The photocurrent (I_m) and photovoltage (V_m) corresponding to the maximum power point (P_m) were then obtained for each cell from the P-V curve. The values of the fill factor (FF) and the cell efficiency (η) were then calculated using:
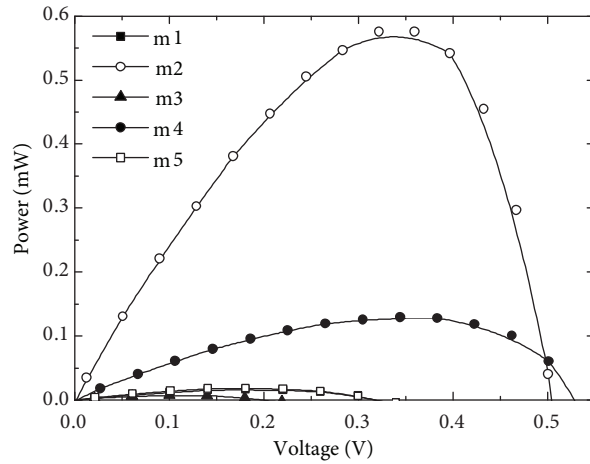
$$FF = \frac{I_m V_m}{I_{sc} V_{oc}}, \tag{1}$$

$$\eta = \frac{FF I_{sc} V_{oc}}{P_{in}}, \tag{2}$$

where I_m and V_m are the photocurrent and photovoltage for maximum power output and I_{sc} and V_{oc} are the short-circuit photocurrent and open-circuit photovoltage, respectively. P_{in} is the power of incident light.
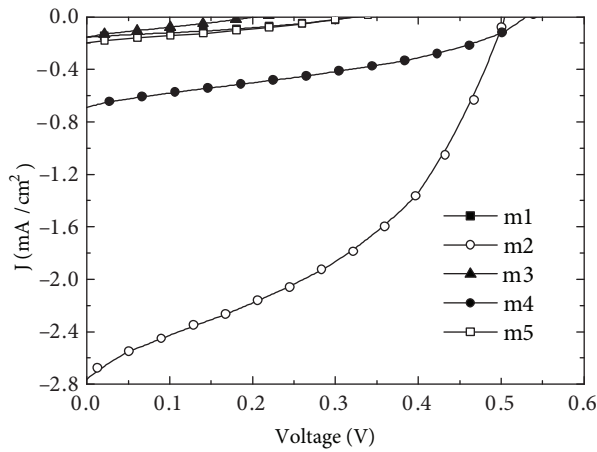
Figure 3. $J - V$ characteristic curves for the fabricated DSSC sensitized using TiO$_2$ nanoparticle as semiconductor.

Figure 4. Power versus voltage curves for all cells.

All results are summarized in the Table. It is clear that the short-circuit current ranged from 0.159 to 2.86 mA/cm^2 for the DSSCs sensitized with 2-((phenylimino)methyl)phenol (m1) and 2-(2-hydroxybenzylideneamino) benzoic acid (m2), respectively. The short-circuit currents obtained for the DSSCs sensitized with 2-(2-hydroxybenzylideneamino)phenol (m3), 4-(2-hydroxybenzylideneamino)benzoic acid (m4), and N-(thiophen-2-ylmethylene)aniline (m5) were, respectively, 0.169, 0.700, and 0.210 mA/cm^2. The open-circuit voltage ranged from 0.218 V for the DSSC sensitized with 2-((phenylimino)methyl)phenol (m1) to 0.562 V for that sensitized with 2-(2-hydroxybenzylideneamino)benzoic acid (m2). Moreover, the DSSC sensitized with 4-(2-hydroxybenzylideneamino)benzoic acid (m4) exhibited a relatively high open-circuit voltage of 0.539 V. Other cells sensitized with 2-(2-hydroxybenzylideneamino)phenol (m3) and N-(thiophen-2-ylmethylene)aniline (m5) showed low open-circuit voltages of 0.2 and 0.3 V. The conversion efficiency of the fabricated cells ranged from 0.007% for the cell sensitized with 2-((phenylimino)methyl)phenol (m1) to 0.575% for that sensitized with 2-(2-hydroxybenzylideneamino)benzoic acid (m2). Therefore, it is clear that 2-(2-hydroxybenzylideneamino)benzoic acid (m2) is a strong candidate as a sensitizer for efficient DSSCs.

Table. Photoelectrochemical parameters of the DSSCs sensitized by various chemical dyes.

Dye	J_{sc} (mA/cm^2)	V_{oc} (V)	J_m (mA/cm^2)	V_m (volt)	P_{max} (mW)	$\eta\%$
m1	0.159	0.218	0.051	0.141	0.007	0.007
m2	2.86	0.562	1.599	0.339	0.575	0.575
m3	0.169	0.219	0.052	0.140	0.0075	0.0075
m4	0.700	0.539	0.333	0.383	0.127	0.127
m5	0.210	0.339	0.102	0.180	0.0185	0.0185

4. Conclusion

Five aldimine derivatives were prepared and examined as sensitizers for efficient dye-sensitized solar cells. A relatively high open-circuit voltage was obtained for the DSSCs sensitized with 2-(2-hydroxybenzylideneamino)benzoic acid (m2) and 4-(2-hydroxybenzylideneamino)benzoic acid (m4). A relatively high short-circuit current was obtained for the DSSC sensitized with 2-(2-hydroxybenzylideneamino)benzoic acid (m2). We suggest that aldimine derivatives such as 2-(2-hydroxybenzylideneamino)benzoic acid (m2) would be strong candidates for fabricating low-cost and efficient dye-sensitized solar cells.

Acknowledgment

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