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New Efficient Organic Compounds In Dye-Sensitized Solar Cells

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Abstract: We demonstrate the use of three new organic photosensitizer compounds in dye sensitized solar cells (DSSCs). One of these compounds is a diazapentadiene derivative [1], while the other two compounds are triazole derivatives [1]. The construction of the cell involved use of a commercially available indium tin oxide (ITO) conductive electrode on which a thin layer of TiO_2 was deposited by chemical vapor deposition (CVD). The other electrode was composed of a thin graphitic layer on ITO. A gelled iodine/iodide combination was used as the redox system.

We have studied the current versus voltage (IV) characteristics and the power versus voltage of these DSSCs at a relatively low light intensity. The efficiency and the fill factor for each cell were consequently calculated.

مركبات عضوية جديدة فعالة للخلايا الشمسية ذات الأصباغ

ملخص: تم استخدام ثلاث مواد عضوية جديدة كمتحسسات ضوئية في الخلايا الشمسية ذات الأصباغ ، أحد هذه المركبات مشتق من مركبات الديزابنتادين و المركبين الأخرين من مشتقات الترايازول.

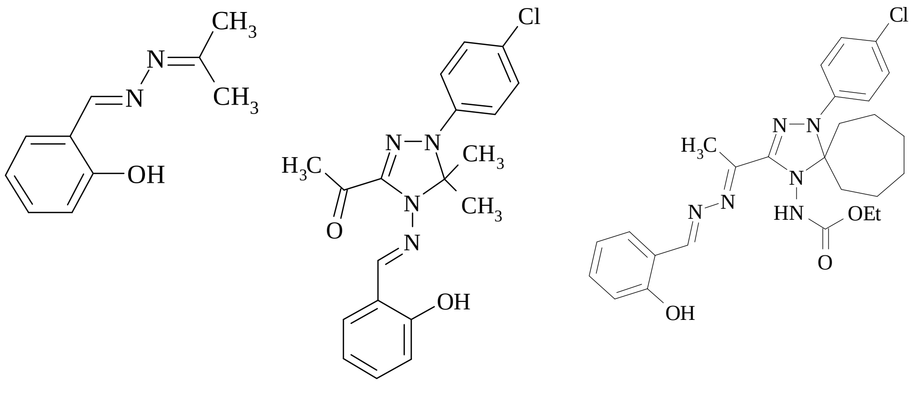
تتركب هذه الخلايا من إلكترودين أحدهما عبارة عن اكسيد القصدير/ الإنديوم(ITO) المرسب عليه طبقة من ثاني اكسيد التيتانيوم بطريقة ترسيب بخارالمادة كيميائيا، أما الإلكترود الأخر فهو عبارة عن طبقة رقيقة من الجرافيت موضوعة على شريحة من ال(ITO)، وقد استخدم وسط جيلاتيني من الأيودين/ أيودايد كعامل مختزل/ مؤكسد.

وقد قمنا بدراسة خواص التيار مع الجهد و القدرة مع الجهد لهذه الخلايا الشمسية تحت إضاءة ضعيفة نسبيا، كذلك تم حساب الكفاءة و معامل الإمتلاء للخلايا.

Introduction

The increasing worldwide energy demand, together with the limited availability of fossil sources and the need to decrease CO_2 production in order to decrease greenhouse gases, has led to an effort to convert solar energy into electricity. Since then, many types of solar cells have been invented [2-8]. Generation of electrical power is achieved by the capability of the photovoltaic device to generate voltage over an external load and current through the load at the same time. One of the most prominent is the dye sensitized solar cell DSSC. The DSSC consists of two electrodes made of a transparent and electrically conducting substrate. A nanocrystalline titanium dioxide TiO_2 layer is deposited on one electrode. A redox, usually I^{-}/I^{-}_{3} electrolyte, fills the space between the two electrodes. The counter electrode is made of a graphitic layer on ITO [6].

An efficient charge transfer sensitizer dye is a prerequisite for developing DSSCs with high efficiency [5,9]. This opens the door to explore DSSCs with different dye molecules. In this article, we report photovoltaic effect from a DSSC solar cell using three new organic compounds (dyes). One of these compounds is a diazapentadiene derivative: 1-(2-Hydroxyphenyl)-4-methyl-2,3-diaza-1,3-pentadiene [1]. The other two compounds are triazole derivatives:3-Acetyl-4-salicylidenamino-5,5-dimethyl-1-(4-chlorophenyl)-1,2,4-triazole, and 3-Acetyl-1-(4-chlorophenyl)-4-ethoxycarbonylamino-1,2,4-triazaspiro[4,6]-undec-2-ene-2-hydroxybenzylidene hydrazone [1]. The structures of these compounds are shown in Fig.1 .



(a) **(b)** **(c)**

Fig. 1. The structures of (a) the diazapentadiene derivative, (b,c), the two triazole derivatives.

Experimental

Transparent conducting electrodes (ITO coated glass substrates purchased from Delta Technologies, USA) were cut into 1 cm x1 cm pieces. The sheet resistance R_s of these electrodes was 8-12 Ω/sq cm.

Undoped TiO_2 porous thin films were deposited by chemical vapor deposition (CVD) method on ITO substrates. Before deposition, the ITO substrates were cleaned by wiping surface with xylene wetted lens cleaning paper followed by drying with a stream of hot air. Then, the cleaned ITO substrates were placed on a pre-heated custom-made hot plate for five minutes at 450 ± 5 ^{o}C in order to achieve thermal equilibrium. The plate's temperature was monitored with a Cromel Alumel thermocouple from 350 ^{o}C to 550 ^{o}C. Fumes of concentrated titanium tetra-chloride ($TiCl_4$), carried by 99.9% oxygen gas, were forced to flow at the rate of 5 l/h through a fine pipette. Chemical vapor deposition (CVD) of TiO_2 films was obtained by allowing the outgoing gas to hit the pre-heated ITO plates for one minute. Finally, the coated samples were allowed to cool down gradually to room temperature. These films were then soaked in a solution containing the dye for overnight.

The counter electrodes were prepared by casting a thin layer of graphite on ITO. The redox electrolyte system used in our work was a gelled iodine/iodide combination. Figure 2 depicts the DSSC cell. The separation between the electrodes was controlled by the standard scotch tape of thickness ~ 40 μm.

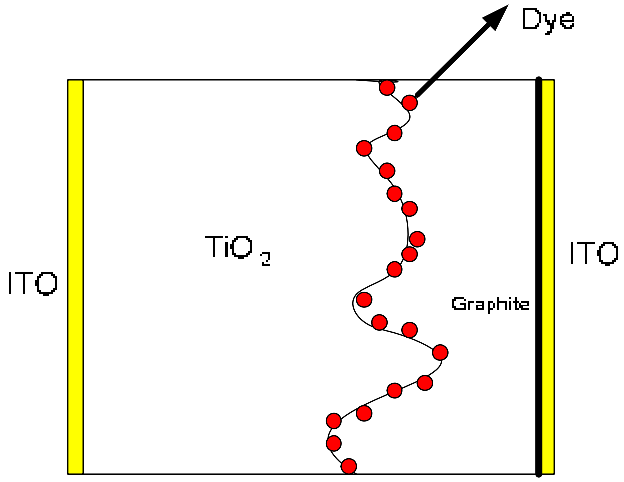


Fig. 2. The DSSC cell

The experimental setup is shown in Fig. 3. A light beam from Revere Automatic (Model no P-808-U) projector with General Electric CZX 500 W lamp was incident normally on the TiO_2 electrode. The light intensity was measured using a Extech instrument light meter model # 401025. The IV characteristics were obtained using a variable resistance. The current data were measured using a picoameter, and the voltage data were measured using hp 3465A digital multimeter. All measurements were conducted at light intensity of about 91watt/m^2, and at room temperature.

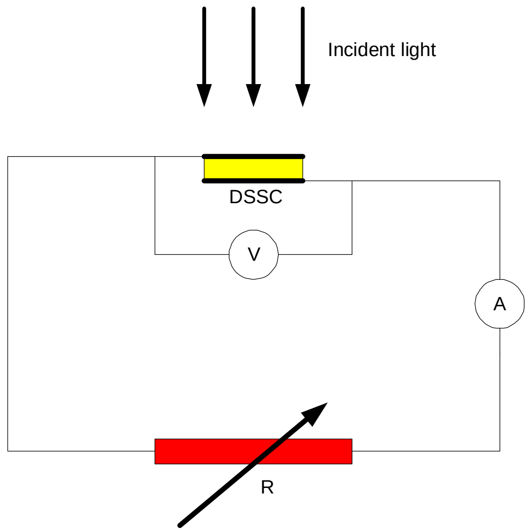


Fig. 3. Experimental setup

Results and Discussion

The energies of the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) of the three compounds, and the energy bandgaps were theoretically computed by first minimizing the energy using AM1 method and a closed shell restricted wavefunction. The HOMO-LUMO energies were deduced from the output of Gaussian 98 software, using hybrid density functional B3LYP and the 6-31G basis set. Results of the HOMO-LUMO energy levels, below vacuum level, for the three dopant molecules and their corresponding bandgaps (Fig.6) are summarized in Table I.

Table I. Theoretical calculation of the HOMO-LUMO energies of the three compounds.

Dye	HOMO	LUMO	Bandgap (eV)
S1	5.206	1.300	3.91
S2	4.728	1.778	2.95
S3	5.338	2.367	2.97

Figure 4 shows plots of the IV characteristics of the three DSSCs under illumination. Measurements were conducted at light intensity of about 91 watt/m^2. Values of the short circuit current I_{sc} and the open circuit voltage V_{oc} were obtained by extrapolating the IV characteristic curves. The power voltage characteristics of the three DSSCs under illumination are illustrated in Fig. 5. It is clear from these two figures that the DSSC with S1 exhibits the highest performance, and the DSSC with S3 exhibits the lowest performance. On the other hand, the DSSC with S2 shows an intermediate performance. We notice that Voc for the DSSC with S3 is a little higher than that of DSSC with S1.

The energy conversion efficiency η of the solar cell defined as the ratio of the output power of the cell and the incident irradiance is given by

$$h = \frac{P_{out}}{P_{in}} = \frac{V_{MP} I_{MP}}{P_{in}} = \frac{V_{oc} I_{sc} FF}{P_{in}}$$

Where FF is called the fill factor defined as the ratio

$$FF = \frac{V_{MP} I_{MP}}{V_{oc} I_{sc}}$$

Values of the short circuit current I_{sc}, open circuit voltage V_{oc}, maximum power current I_{MP}, maximum power voltage V_{MP}, fill factor FF, and efficiency η are shown in Table II.

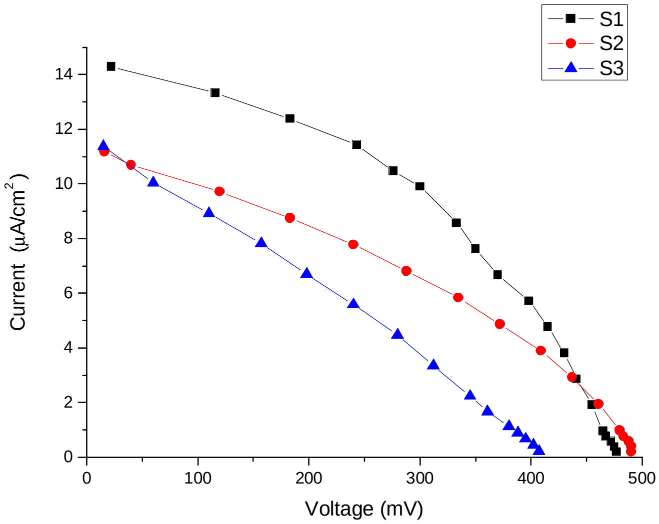


Figure 4. IV characteristics of the three DSSCs under illumination.

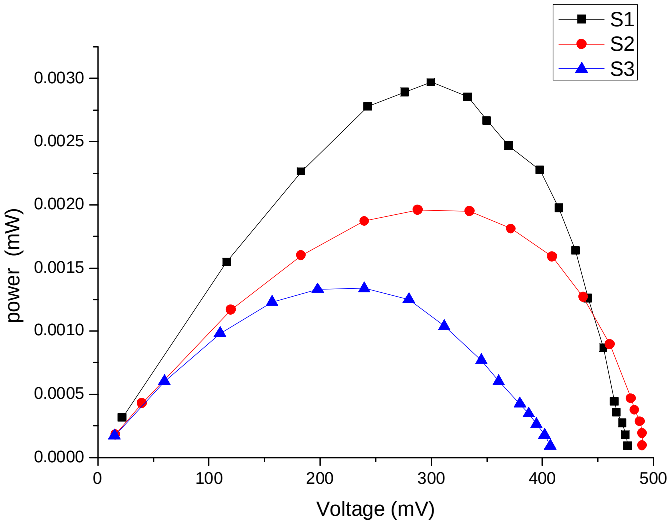


Figure 5. The power voltage characteristics of the three DSSCs under illumination.

Table II. Values of I_{sc}, V_{oc}, I_{MP}, V_{MP}, FF, and η of the three DSSCs at about 91 watt/m^2.

DSSC	I_{sc} (µA)	V_{oc} (mV)	I_{MP} (µA)	V_{MP} (mV)	FF	η %
S1	14.3	477	9.9	300	0.44	0.033
S2	11.2	490	7.8	288	0.41	0.025
S3	11.4	407	5.6	240	0.29	0.015

The principle of operation and energy level scheme of the DSSC is illustrated in Fig. 6. Photo-excitation of the sensitizer (S) is followed by electron injection into the conduction band of the oxide semiconductor. The dye molecule is regenerated by the redox system, which itself is regenerated at the counter electrode by electrons passing through the external load. The maximum theoretical voltage corresponds to the difference between the redox potential of the mediator and the Fermi level of the mesoporous film. Details of this operation can be found elsewhere [5, 10-12].

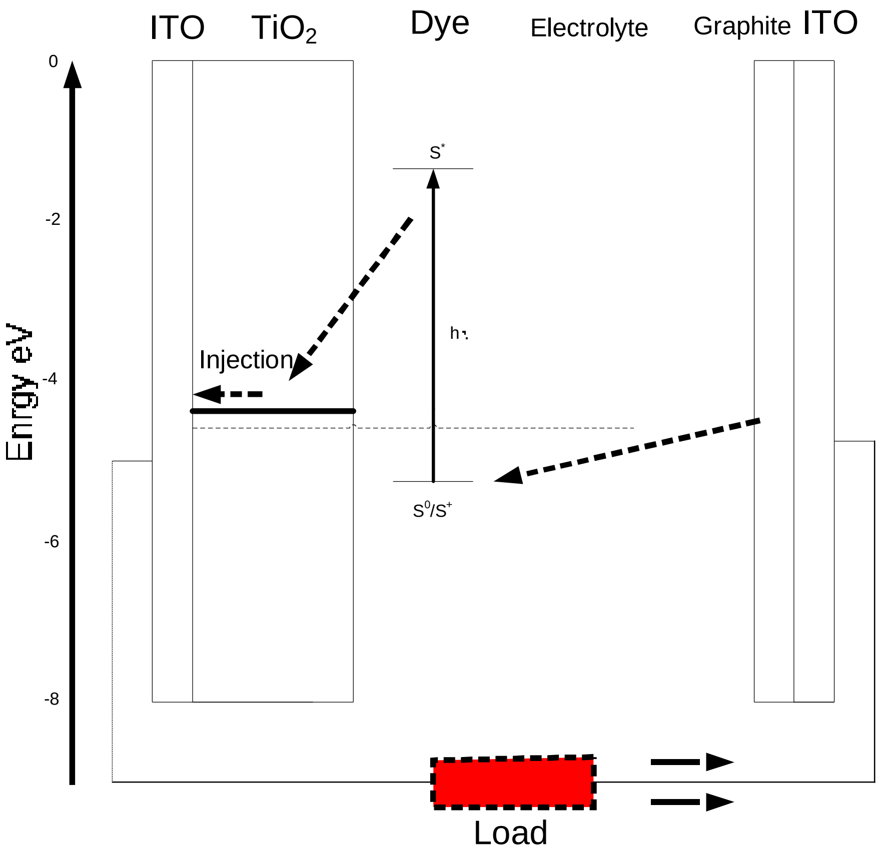


Fig. 6. A schematic presentation of the operating principles of the DSSC (Dye S1).

It is well known that the efficiency of any photoconverter is dependent on the incident light intensity[13].Furthermore, varying the light intensity incident on a solar cell changes all solar cell parameters, including the short-circuit current, the open-circuit voltage, the efficiency, the FF, and the impact of series and shunt resistances. Low values of the fill factors (FF) for the DSSCs are limited in general by the series resistance losses, non-ideal dark diode currents, light intensity dependent recombination, and sometimes, due to shunt resistance losses [14].

The low values of η in our samples may be due to the faster charge recombination losses resulting directly from the increased electron mobility. [14], or it may be due to the relatively weak irradiance incident on the cell. This decrease in η at low incident light intensities can be explained by the loss of photogenerated electrons via surface-state-mediated electron transfer into the electrolyte. The strong dependence of the incident photon energy to current conversion efficiency on the incident light intensity has consequences for the efficiency of a dye-sensitized solar cell at low illumination levels [15]. On the other hand, comparing the energy levels for cells with S2 and S3 suggests that S3 should result in better cell performance. However, experimental results show a different behavior where the efficiency for the cell with S2 was higher than that with S3. It is interesting to indicate that this behavior could easily be interpreted by looking at the total strain of the dopant molecules , which in part presents the planarity of the double bond system. This planarity is essential for free flow of electrons through the system. Another factor that could be considered is the van der Waals forces between molecules which can lead to aggregation possibility. Molecules with high forces tend to aggregate, thus forbidding free absorption of energy and transfer of electrons to TiO_2. Comparison between the three dopant molecules is presented in Table III (calculated using Molecular Modelling Pro from Chemsw).

Table III. Comparison of dopant molecules in terms of total strain and van der Waals forces.

Dye	Total Strain (kcal/mol)	Van der Waals Energy
S1	7017.4	6255.6
S2	28788.9	26766.4
S3	101951.3	99157.2

It is evident from the results presented in the table above that the total strain and van der Waals forces for S3 are much higher than that calculated for S2, and both are much higher than that shown for S1. This predicts better performance of S1 than S2 or S3. At the same time results also suggest that S3 should show the lowest efficiency of the three dopants, which was experimentally observed.

Conclusion

In this article, we demonstrate the use of three new organic compounds (dyes) as efficient Materials in dye sensitized solar cells (DSSC). One of these compounds is a diazapentadiene derivative, while the others are triazole derivatives. Also, a gelled I^-/I^-_3 electrolyte combination was used as the redox system. The HOMO-LUMO energies of the three compounds were calculated from the output of Gaussian 98 software using hybrid density functional B3LYP and the 6-31G basis set.

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