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Photovoltaic properties of polymer based organic solar cells adapted for non-transparent substrates

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ABSTRACT

This paper explains a semi-transparent photovoltaic device structure using polymer based materials for light harvesting. Poly(3-hexylthiophene-2,5-diyl):[6,6]-phenyl C_{61} butyric acid methyl ester (P3HT:PCBM) and poly(2-methoxy-5-(3′,7′-dimethyloctyloxy)-1,4-phenylenevinylene) (MDMO-PPV:PCBM) as photoactive nano-layers were utilized and semi-transparent cells were compared with reference cells. Photoelectrical properties of developed devices were investigated. Also influencing factors of power conversion efficiency of devices were determined and possible application areas including solar harvesting textiles were discussed.

1. Introduction

Renewable energies including solar cells attract considerable attention due to the inevitable end of fossil fuels and due to global warming and other environmental problems they caused, in recent years. To obtain clean and renewable energy, total photovoltaic technology capacity in the world exceeded 9 GWP at the end of 2007 [1]. Silicon based solar cells, photochemical solar cells and organic solar cells are some of the developing photovoltaic technologies. Among the solar cell technologies, traditional silicon based solar cells which have been used in space and military applications for a long time still dominate in the photovoltaic markets. Today, the silicon based solar cells have ca. 25% power conversion efficiency in laboratory conditions [2] and often take place in human daily life and industrial applications to produce electricity. However, they have some disadvantages such as using expensive materials and manufacturing techniques. Also, silicon based photovoltaics are generally rigid and flat that is why application fields are limited. Ongoing efforts focus on for reducing the fabrication cost of solar cells and for improving the efficiencies of existing devices. In this respect, an organic solar cell using organic semiconductors which are cheaper than inorganic semiconductors and can be synthesized with easier processes is a promising approach owing to its unique features including low-cost, flexibility and lightness for solar energy conversion. Additionally, their chemical and physical properties can be changed or tuned easier [3,4]. The power conversion efficiency under AM1.5 conditions for polymer based solar cells has just exceeded 5% in laboratory conditions [2].

A solar cell is a device which converts energy of photons into electrical energy. The photovoltaic effect requires the generation of electron–hole pairs in the device under illumination and their subsequent collection at reverse electrodes. The most widely used technique for manufacturing the polymer based organic solar cells is based on blend of electron-donating and electron-accepting organic materials [4]. Organic solar cells can be fabricated using various techniques including solution processing, vacuum evaporation [3,4], printing [5–8] or nanofibre formation [9] and electrospinning techniques [10] at ambient conditions. Organic solar cells can be developed on various types of substrates such as glass, plastic foil [4,11], optical fibers [12–14], etc.

The studies about (semi-)transparent upper electrode for organic solar cells [15–23] show that (semi-)transparent solar cells can be used for manufacturing tandem solar cells and multiple devices to achieve high device performance. Also, (semi-)transparent metal layers can create innovative research areas for developing different solar harvesting structures such as solar harvesting windows [24] or textiles [25,26].

This study was investigated to demonstrate the effect of semi-transparent cathode and illumination direction to organic solar cell

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performance. Our aim is also develop a solar cell structure which can be applied onto non-transparent and flexible substrates like textiles. Well-known solar cell materials were used and modified, and then the photovoltaic properties of these conventional and modified solar cells were presented using experimental results.

2. Experimental details

A polymer based organic solar cell (see Fig. 1) generally consists of a transparent conductive bottom electrode like indium tin oxide (ITO) obtained generally coated on flat substrates like glass, in the photovoltaic market. A poly(3,4-ethylenedioxythiophene):poly (styrenesulfonate) (PEDOT:PSS) layer is coated on top of ITO layer to facilitate charge injection and extraction. The light absorbing organic layer is placed between anode and cathode. This nano-composite layer is generally formed by blending a fullerene derivative and conjugated polymer. The upper electrode can be generally formed by vacuum deposition of low work function metals (aluminum (Al), calcium (Ca), etc.) onto photoactive layer. Lithium fluoride (LiF) is the interfacial layer between active layer and cathode and also used in organic photovoltaics to enhance electron injection and to reduce the contact resistance [27,28]. Aluminum is a common metal electrode used in photovoltaics to collect the electrons. Additionally, solar cells used outdoors are generally constructed on flat and usually on rigid surfaces.

2.1. Preparation of solar cells

In this study, four different types of solar cells were produced. Two of them as reference cells have ca. 100 nm thick LiF/Al and others have ca. 10 nm thick LiF/Al layer. Solar cells consisting of thinner cathode were measured from two different directions (from lower and upper electrodes, respectively) and so, six different results were obtained from photoelectrical experiments of devices (see Table 1).

The polymer based organic solar cells were prepared by the following procedures:

Conventional ITO coated glasses (Merck) with a specific ITO thickness (ca.120 nm) and sheet resistance (less than 15 Ω/cm^2) were cut in $15 \times 15\ mm^2$ to be used in experimental studies. For etching some part of the ITO layer from the glass substrate, an acid mixture containing hydro chloric acid (HCl), nitric acid (HNO_3) and water (H_2O) were applied for 30 min. After etching process, samples were washed with a detergent (helmanex in water), acetone, iso-propanol, ethanol, methanol and water in ultrasonic bath for 15 min, respectively. Later, an aqueous solution of highly conductive PEDOT:PSS (Bayer AG, Baytron PH 500) mixture consisting of dimethyl sulfoxide (DMSO) to increase the conductivity

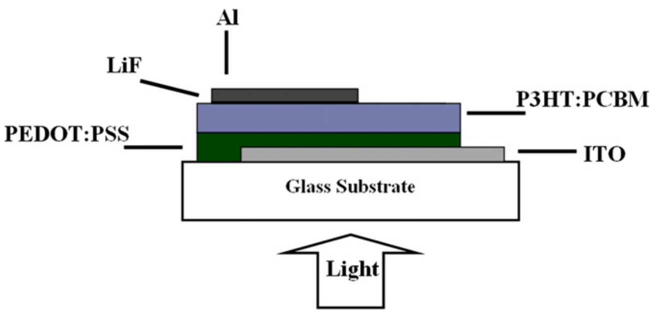

Fig. 1. Schematic demonstration for illumination of conventional P3HT:PCBM based organic solar cell through anode (Experimental device set up and measurement conditions are same for MDMO-PPV:PCBM based solar cells).

Table 1
Device structures and illumination directions.

Device	Solar cell structure	Illumination side
a	Glass/ITO/PEDOT:PSS/P3HT:PCBM/LiF/Al (~100 nm)	Anode
b	Glass/ITO/PEDOT:PSS/P3HT:PCBM/LiF/Al (~10 nm)	Anode
c	Glass/ITO/PEDOT:PSS/P3HT:PCBM/LiF/Al (~10 nm)	Cathode
d	Glass/ITO/PEDOT:PSS/MDMO-PPV:PCBM/LiF/Al (100 nm)	Anode
e	Glass/ITO/PEDOT:PSS/MDMO-PPV:PCBM/LiF/Al (10 nm)	Anode
f	Glass/ITO/PEDOT:PSS/MDMO-PPV:PCBM/LiF/Al (10 nm)	Cathode

was prepared and spin-coated (1500 rpm and 70 s) onto pre-cleaned ITO coated glass substrates. Then, samples were completely dried for 5 min at 100 °C on hot plate. The resulting PEDOT:PSS film thickness is ca. 200 nm. Chemical structure of PEDOT:PSS is shown in Fig. 2.

Light absorbing layer was deposited by spin-coating technique on top of PEDOT:PSS covered on top the ITO coated substrates for 70 s at 1500 rpm. Two kinds of photoactive material mixtures were prepared with following procedure: Firstly, P3HT:PCBM mixture was dissolved in chlorobenzene with a ratio of 1:0.8 (w/w) and stirred for 24 h at room temperature. Meanwhile, MDMO-PPV: PCBM mixture with a ratio of 1:4 (w/w) was prepared by dissolving in chlorobenzene, and then stirred for 24 h at 40 °C. The resulting photoactive film thickness is ca. 200 nm. P3HT (Rieke Specialty polymers) and MDMO-PPV (Covion Merck) worked as electron-donating polymers and PCBM (SOLENNE) worked as electron-accepting material in the blend. Chemical structures of P3HT, MDMO-PPV and PCBM are shown in Fig. 2, respectively. Subsequently a ca. 100 nm thick Al/LiF contact was evaporated thermally under vacuum (10^{-6} mbar) on top of the photoactive layers for reference (conventional) cells. However, top metal electrode consisting of 0.7 nm LiF and 10 nm Al was deposited with same technique on photoactive layers to obtain modified organic solar cells. The schematic demonstration of both types of organic solar cells using polymer based nano-layers is given in Fig. 3. Later, silver paint was put onto the electrodes of the solar cells to take better conductivity from contacts.

2.2. Photovoltaic characterization of the cells

The photovoltaic performances of conventional and modified organic solar cells were tested in a nitrogen environment inside a glove box system (MBraun) at room temperature. A Keithley 236 source measure unit was used to measure current density versus voltage ($I-V$) characteristics of the solar cells in the dark and under light obtained with a calibrated solar simulator source (K.H. Steuernagel Lichttechnik GmbH) providing AM1.5 conditions (illumination intensity of 100 mW/cm^2). All four devices were illuminated through ITO coated glass, conventionally. Later modified organic solar cells having 10 nm cathode were illuminated from semi-transparent top electrode (cathode) side (reference cells (conventional organic solar cells) were only illuminated from ITO coated glass (anode) direction for comparison).

The photovoltaic power conversion efficiency (η) of an organic solar cell is calculated by the following Eq. (1);

$$\eta = \frac{I_{sc} \times V_{oc} \times FF}{P_{in}} \tag{1}$$

Where I_{sc} is the short-circuit current and V_{oc} is the open-circuit voltage in the system. Maximum power (P_{max}) is calculated by Eq. (2) and fill factor (FF) of the solar cell is determined by Eq. (3);

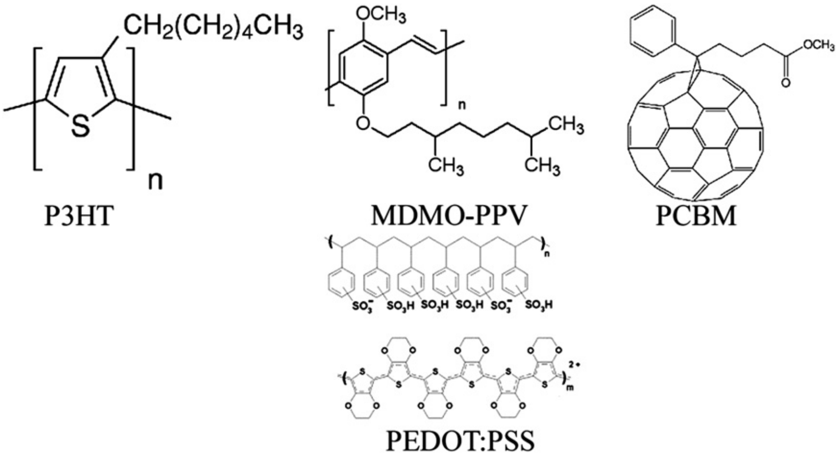

Fig. 2. Chemical structures of P3HT, MDMO-PPV, PCBM and PEDOT:PSS.

$$P_{max} = I_{mpp} \times V_{mpp} \quad (2)$$

$$FF = \frac{I_{mpp} \times V_{mpp}}{I_{sc} \times V_{oc}} \quad (3)$$

Division of P_{max} by the product of I_{sc} and V_{oc} gives the fill factor FF. I_{mpp} and V_{mpp} are the current and the voltage at the maximum power point of an I–V curve in the fourth quadrant. P_{in} is the incident light power density [3].

Ultraviolet–visible transmission spectra of thin films were measured using Varian Cary 3G UV–visible spectrophotometer. Spin-coating technique was used to deposit P3HT:PCBM and MDMO-PPV:PCBM mixtures on microscope glasses and thermal evaporation technique was utilized for forming LiF/Al thin metal layer on microscope glass.

3. Results and discussions

Generally, an organic solar cell is developed on transparent substrates like glass and uses transparent anodes such as indium-doped tin oxide (ITO) and fluorine-doped tin oxide (FTO) so that light can enter inside of the photovoltaic device (see Figs. 3 and 4). In this study, polymer based solar cells were modified using a transparent electrode as cathode. Contrary to conventional solar cells using 100 nm metal upper electrode and non-transparent modified solar cells consisted of very thin (ca. 10 nm) upper electrode allowing light transmittance. The photoelectrical properties and power conversion efficiency of devices were compared to demonstrate their possible usage fields. Active areas of solar cells were between 6 and 14 mm^2 and 15 devices were prepared and measured for each design.

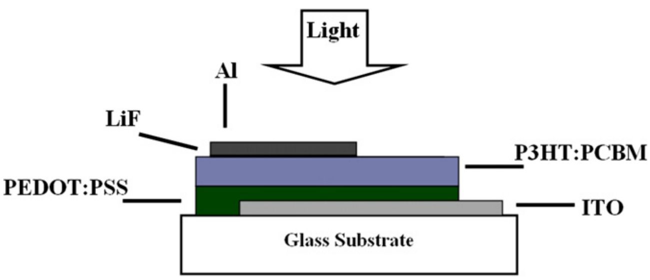

Fig. 3. Schematic demonstration for illumination of modified P3HT:PCBM based organic solar cells through semi-transparent cathode (Experimental device set up and measurement conditions are same for MDMO-PPV:PCBM based solar cells).

While a 100 nm thick LiF/Al layer was non-transparent, 30% semi-transparency was obtained using 10 nm of LiF/Al, in visible range (see Fig. 5). However, a 120 nm thick ITO layer is almost transparent (90%). Due to ITO material is expensive and brittle and has difficult process techniques to be used on flexible substrates, semi-transparent metal layer which can overcome these disadvantages was considered, here. Also, the transmission of the metal layer can be significantly enhanced by the right architecture and combinations, in future studies.

The AM1.5G spectrum shows that the sun emits approximately 26% of the photons in the visible part of the spectrum. [24]. The active layer transmission of devices based on P3HT:PCBM and MDMO-PPV:PCBM is depicted in Fig. 6. Devices of this kind using PCBM as acceptor and P3HT and MDMO-PPV as donor have been presented previously [3,4]. They are well-known materials and have the advantage that is to absorb most of the photons in visible range of sunlight. Further optimization of the devices might include additional materials which can absorb light in wide range of solar spectrum to increase the power conversion efficiencies of devices.

The photovoltaic characteristics of conventional and modified organic solar cells consisting of P3HT:PCBM and MDMO-PPV:PCBM

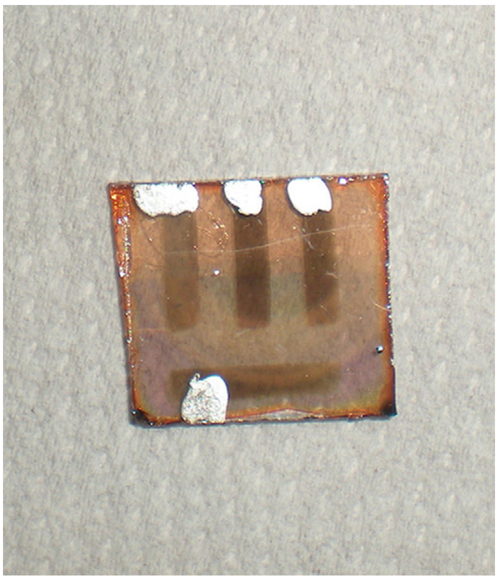

Fig. 4. Photograph of a semi-transparent polymer based solar cell.

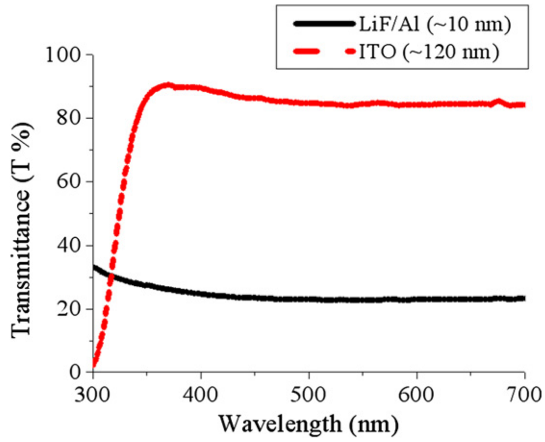

Fig. 5. The comparison of ITO (120 nm) layer and LiF(0.7 nm)/Al (10 nm) metal layer on general transmittance spectra.

light absorbing layers are given in Table 2. Current density versus voltage (I–V) characteristics of the devices are also demonstrated in Fig. 7 in terms of illumination directions and cathode thicknesses at room temperature.

As can be seen from Table 2 and Fig. 7, a decrease in short-circuit current density is seen in modified cells in comparison with results of reference cells' measurements using same illumination direction. This is mainly due to the reduced thickness of the top electrode to provide semi-transparency. However, with this modified design, device can be illuminated from both bottom and top.

When reference cells and modified cells illuminating through anode were compared, it was seen that efficiency decreased in both devices. Loss in efficiency of MDMO-PPV:PCBM based devices was ca. 50% and 85% loss was occurred P3HT:PCBM based solar cells. A thinner (10 nm) top electrode in the device design caused a significant decrease in short-circuit current density (I_{sc}) and power conversion efficiency (η). There is a trade-off between electron collecting ability and metal cathode thickness.

Photovoltaic characteristics including largest maximum power output (P_{max}) and voltage and current at maximum power point of linear I–V curves of modified devices are demonstrated in Fig. 8 and obtained results were presented in Table 3. The intersections with the horizontal and vertical axis of linear I–V curves are the open-circuit voltage (V_{oc}) and the short-circuit current (I_{sc}), respectively. Results for maximum power output calculations of devices show that MDMO-PPV:PCBM based modified solar cell gave higher

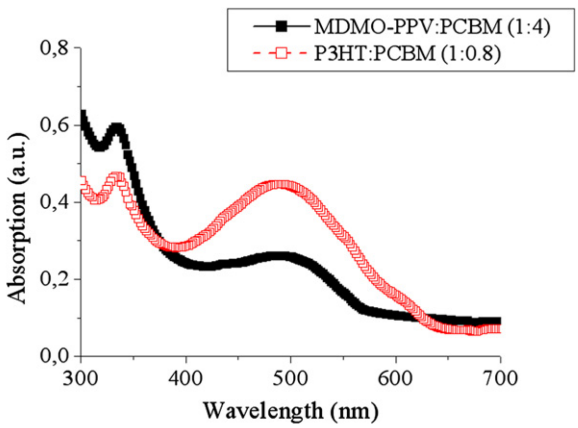

Fig. 6. General absorption spectra of P3HT:PCBM and MDMO-PPV:PCBM thin layers on glass.

Table 2
Photoelectrical results of the organic solar cells applied on different illumination directions.

Device	I_{sc} (mA/cm^2)	V_{oc} (mV)	FF (%)	η (%)
a	8.75	650	50.2	2.86
b	2.71	660	34.4	0.62
c	1.13	620	36.1	0.25
d	5.31	650	32.1	1.11
e	1.85	780	37.3	0.53
f	1.35	740	37.7	0.37

current and voltage at its maximum power point than P3HT:PCBM based modified devices.

When modified solar cells were illuminated through cathode, it was seen that efficiency decreased in both devices, again. Efficiency loss was occurred ca. 30% in MDMO-PPV:PCBM based solar cells and 60% in P3HT:PCBM based solar cells. It was due to both transmission and absorption difference between ITO and LiF/Al layers. Transmission of ~10 nm LiF/Al layer was lower than ITO layer ca. 55% in visible wavelengths. Number of photons entering into device influences the generation of electron–hole pairs in photoactive layer. Although there was a slightly decrease in short-circuit current density (I_{sc}) and power conversion efficiency (η) between modified solar cells illuminating from different directions, moderate efficiency which can be a promising first step for developing wearable photovoltaics was obtained in Device c and Device f.

As can be seen from Table 2 and also from Fig. 7, P3HT:PCBM based conventional solar cells showed higher I_{sc} and efficiency results than MDMO-PPV:PCBM based solar cells for reference cells.

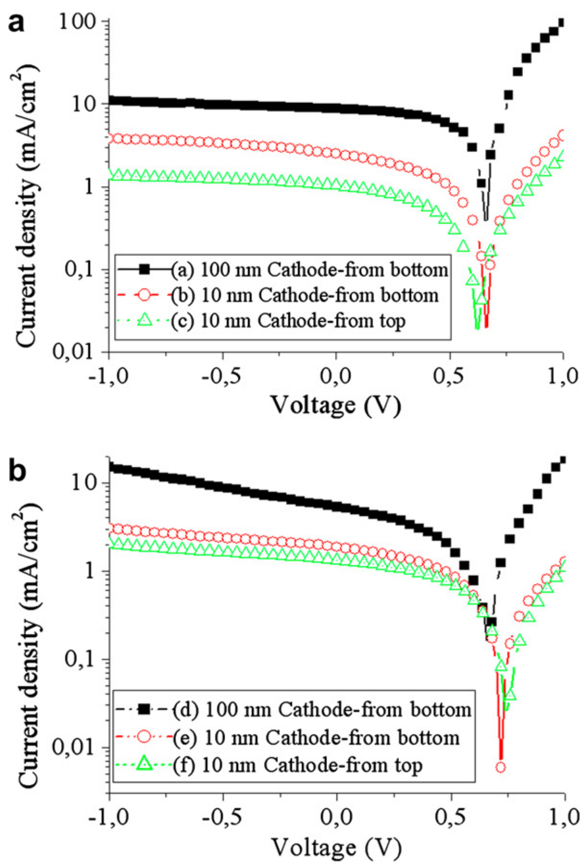

Fig. 7. Semi-logarithmic I–V curves of P3HT:PCBM (Devices: a, b and c) (**a**) and MDMO-PPV:PCBM (Devices: d, e and f) (**b**) based organic solar cells applied from different illumination directions.

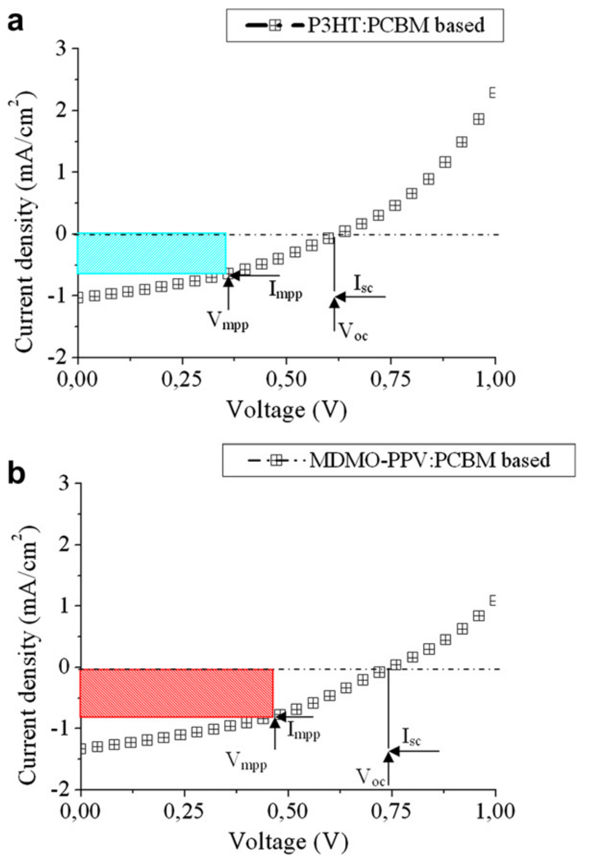

Fig. 8. Linear current density versus voltage (*I*–*V*) curves of the organic solar cells using semi-transparent cathode and illuminating through cathode (P3HT:PCBM (**a**) and MDMO-PPV:PCBM (**b**)).

When thinner cathode was used, it was reverse. The highest power conversion efficiency of modified solar cells was 0.37% in MDMO-PPV:PCBM based solar cells (Device f) illuminating through top contact.

The photoelectrical characteristics of solar cells, especially voltages and current densities are better in conventional solar cells having 100 nm of cathode than modified solar cells having 10 nm when illuminated from glass side, in Table 2. Due to graded transparency of cathodes and due to contacts were damaged after several photoelectrical measurements, an optimization is needed for developing more stable devices. Measurement technique was also changed according to modified solar cells illuminating through upper electrode. For this reason, solar cells were turned down to take the light into the device. However, in this situation, mechanical protection must be somehow used if not contacts or photoactive layer could be damaged.

Such a solar cell may be used in windows (power generating windows) [17,24,29] after applying an appropriate protective layer. However, metal top electrode and organic photoactive layers are not stable in ambient conditions and can be oxidized when exposed to air. For the production of polymer based organic solar cells, difficulties [30,31] including photochemical degradation of polymers and electrodes and also low power conversion efficiency must be overcome. To achieve higher device performance, efficient absorption of solar spectrum is one of the major concerns. Therefore, subjects of using novel upper electrodes, different metal combinations, low band gap materials and non-vacuum coating techniques [8] studied by researchers, recently, will be considered for future studies. Solar cells having graded or completely transparent cathodes will be also useful for architectural works with diverse forms on non-transparent substrates such as textiles to obtain wearable and portable photovoltaics.

Table 3
Photovoltaic characteristics of modified organic solar cells.

Devices	V_{mpp} (V)	I_{mpp} (mA/cm^2)	P_{max} (W)
(c) P3HT:PCBM based	0.36	0.64	2.31
(f) MDMO-PPV:PCBM based	0.46	0.81	3.71

4. Conclusions

This study investigated the optical and photovoltaic properties of polymer based solar cells having semi-transparent cathodes. Our aim was to use a semi-transparent upper electrode for allowing light transmission into the device and find an applicable approach for textiles. Although magnitudes of short-circuit current density and power conversion efficiencies of modified solar cells were low for practical applications, semi-transparent upper electrode was successfully fabricated onto photovoltaic device. It has to be mentioned that organic solar cells have low efficiency, today, comparing to the inorganic solar cells. However, novel advanced materials and appropriate application methods can enhance the new device structures and efficiencies. To obtain the light transmission into the solar cell by lowering the cathode thicknesses, can be a useful approach and can be improved with future studies on this case. Such an optimized and adapted (semi-) transparent solar cell can open up new application areas of photovoltaics in textiles.

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