Effect of Small-molecule Sensitizer on the Performance of Polymer Solar Cell

Yasser A. M. Ismail^{1*}, Tetsuo Soga² and Takashi Jimbo³

¹Department of Physics, Faculty of Science, Al-Azhar University, Assiut 71524, Egypt.
²Department of Frontier Materials, Nagoya Institute of Technology, Nagoya 466-8555, Japan.
³Research Center for New-Device and System, Nagoya Institute of Technology, Nagoya 466-8555, Japan.
*Author to whom correspondence: <u>Yasser ami@yahoo.com</u>

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Abstract: For improving optical absorption of organic solar cells, a small-molecule sensitizer, as a third material, is usually incorporated into binary solar cell system. In many cases, light harvesting can be improved but on the expense of charge carrier mobility of the solar cells. This obstacle can be addressed through the understanding solar cell physics. In the present work, we try to deeply understand the physics of the long-chain polymer solar cell composed of poly(3hexylthiophene) (P3HT), as a donor polymer, and [6,6]-phenyl- C_{61} butyric acid methyl ester (PCBM), as an acceptor molecule. This understanding can be acquired through the effect of coumarin 6 dye (C6), as a small-molecule sensitizer, on optical absorption and photocurrent of the most common solar cell. From optical spectroscopy we found that, the C6 dye, as a small molecule, did not vary conjugation length of the long-chain polymer in the P3HT: PCBM: C6 solar cell. This was indicated from (1) unchanged vibronic structure of the P3HT after adding C6 dye and from (2) matching in the wavelength between absorption peaks of both pristine C6 and P3HT after adding C6 dye into blend. From photocurrent spectroscopy we found that, the incorporation of C6 dye, as a sensitizer, into P3HT: PCBM binary contributed to photocurrent and formed an additional charge carrier generation site through the C6: PCBM combination, which was individually found among with P3HT:PCBM combination in the same solar cell. In the same time, the C6 dye, as a shortchain molecule, restricted the transport of charge carriers generated by P3HT as a result of low hole mobility of the C6 short-chain molecules. Through the present study, the incorporation of a small-molecule sensitizer into polymer solar cell may acquire better understanding for the performance of the most common P3HT: PCBM solar cell.

Keywords: Polymer Solar Cells; Device Physics; P3HT: PCBM Solar Cell.

Introduction

Organic photovoltaics (OPVs) are solar cells that employ organic materials, either polymers (macromolecules) or small molecules, to absorb light and produce free electrons. Interest in OPVs began in the 1990s when Sariciftci et al. [1] demonstrated photo-induced charge transfer between organic molecules in 1992. Over the last two decades, OPVs have garnered serious attention because they have the potential to be an economically viable source of renewable energy. Polymers can be produced cheaply, and since plastics have a high optical absorption coefficient, very little material is needed [2]. In addition, polymers can be dissolved in solvents and deposited on substrates using wetprocessing techniques such as spin coating or roll-to-roll (R2R) printing. Solution processing makes large-scale, low-cost production a possibility. Organic solar cells are also attractive because they lack a rigid crystalline lattice and can be deposited on flexible substrates.

Bulk heterojunction (BHJ) polymer solar cell based on poly(3-hexylthiophene) (P3HT) (which is the hole-transporter and donor material) and [6,6]-phenyl- C_{61} butyric acid methyl ester (PCBM) (which is the electron-transporter and acceptor material) is the largest in researchers' investigations and studies for improving its power conversion efficiency [3]. This device offers considerable promise for use in new solar energy technologies due to its flexible material properties and potential for low-cost manufacture [4].

The reported efficiencies for organic solar cells have risen sharply, from the rather modest 1% in the first BHJ device in 1995 [5]. By 2003, P3HT:PCBM cells with 3.5% were reported [6], and in 2007 multiple groups reported efficiencies of 4- 4.5% with this material combination [7-10].

Organic molecular and polymeric semiconductors can form films with complex morphologies and varying degrees of order and packing modes through the interplay of a variety of non-covalent interactions. The molecular structure of the organic polymers consistently presents a backbone along which the carbon (or nitrogen, oxygen, sulfur) atoms are sp²-hybridized and thus possess a π -atomic orbital. The conjugation (overlap) of these π orbitals along the backbone results in the formation of delocalized π molecular orbitals, which define the frontier molecular electronic levels (highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO)) and, therefore, determine the optical and electrical properties of the (macro) molecules. The overlap of the frontier π molecular orbitals between adjacent polymer chains characterizes the strength of the intermolecular electronic couplings, which represent the key parameter governing charge carrier mobilities. One must distinguish between intramolecular charge transport along a conjugated polymer chain and intermolecular charge transport between adjacent polymer chains. The former, which is specific to conjugated polymers, is the most efficient. Therefore, charge carrier mobilities and, consequently, performance of polymer solar cells strongly depend on morphology of the solar cell active layer.

On the other hand, there are many attempts for improving the performance of polymer solar cells by improvement light harvesting through the adding a dye sensitizer, as a third material included in the soar cell active layer [4,11]. In such case, light harvesting of the solar cell active layer can be improved, but on the expense of charge carrier mobility, which may largely vary depending on molecular structure and conjugation of the sensitizer. So that, for solving this problem, the investigators are have to understand device physics of the polymer solar cells more deeply for improving light harvesting without decreasing charge carrier mobility and whole performance of the cells. For more understanding solar cell physics, we need to illustrate and examine the behavior of a solar cell composed of highly conjugated polymer, such as P3HT, when a small-molecule sensitizer, as a third material, is incorporated in the solar cell active layer. In this case, the small molecule contributes to light harvesting as a sensitizer, and, in the same time, itself is the responsible for enhancing or reducing the charge carrier mobility, and therefore, performance of the polymer solar cells, depending on its ability for transferring charge carriers in the solar cells.

As an attempt for understanding physics of polymer solar cells, we use in this work one of the coumarin dyes, as a small-molecule sensitizer, which can be incorporated as a third material in the BHJ blend of the P3HT:PCBM binary. The coumarin dyes have been used as light absorbers for solar cell applications [12], and also they have been extensively used as photosensitizers in Grätzel-type dye-sensitized solar cells [13,14], as dopants in polymer light emitting diodes (LEDs) [15,16], and as luminescent solar concentrators applied upon solar cells [17,18]. Because of its light harvesting property accompanied with its energetically favorable incorporation into P3HT: PCBM binary, we choose coumarin 6 dye (C6) to study its impact on light harvesting, as a sensitizer, and performance of one of the most common polymer solar cells.

When selecting the donor and acceptor material pair for photovoltaic application, one needs to consider their energy levels. The HOMO and LUMO of the material pair need to match in such a way that, exciton dissociation (charge separation) will be favored at the interface of the two materials. The C6 dye in combination with the P3HT: PCBM binary shows a thermodynamically suitable charge separation at the C6/PCBM interface, as shown in Fig. 1 from references [19] and [20].

After absorption of light by the sensitizer C6 dye, the charge separation at the C6/PCBM interface can occur as a result of decreasing in electron affinity (higher LUMO) and ionization potential (higher HOMO) of the C6, as a donor, in comparison with those of PCBM, as an acceptor. Therefore, the interfacial electric field at the C6/PCBM interface drives charge separation.

The generated electrons are transported through PCBM into the cathode, while the generated holes are transported either from C6 directly towards anode or from C6 into anode via P3HT. Therefore, the C6 dye can be used in solar cells for light harvesting and electron donation in combination with the P3HT: PCBM binary.



Fig. 1. Energy-level diagram of C6 dye (from reference 19) and the rest components (from reference 20).

In the present work, we try to understand the physics of the most common solar cell binary composed of P3HT, as a donor polymer, and PCBM, as an acceptor molecule, through the effect of C6 dye, as a small-molecule sensitizer, on light harvesting and photocurrent of the cell. We have constructed indiumtin-oxide Polv (3,4-ethylenedioxythiophene) (styrenesulfonate) (ITO) poly (PEDOT:PSS)/P3HT:PCBM:C6/Al BHJ solar cells having the composition 1:2 weight percentage (wt.%) of the P3HT:PCBM binary with increasing C6 dye concentration in the solar cell active layer from 0.2 up to 5 mg. The present study may increase our knowledge about behavior of long-chain polymer solar cell when a small-molecule sensitizer, as a third material, is blended within it. The optical absorption spectroscopy and photocurrent spectroscopy under AM1.5 white light illumination with an intensity of 100 mW/cm² are investigated for solar cells with varying C6 dye concentration in solar cell active layer.

Experimental procedures Materials and solutions

The C6 or [3-(2-benzothiazolyl)-7-(diethylamino)coumarin] dye (99%, Sigma-Aldrich), regioregular P3HT and PCBM (99.2%, Frontier Carbon Corporation, Japan) were used without further purification.

The C6 dye was added with different amounts as 0.2, 0.5, 1 and 5 mg into 30 mg/mL of P3HT: PCBM (1:2 wt. %) blend using 1,2-dichlorobenzene (Tokyo Chemical Industry Co. Ltd, Japan) as a solvent. The blend solutions were vigorously stirred for more than 24 h at room temperature under nitrogen atmosphere in a glove box to maximize mixing while avoiding touching the vial cap.

Film and device fabrication

For organic thin film fabrication, the P3HT:PCBM and P3HT:PCBM:C6 films (with different concentration of the dye in the blends) were prepared by spin-coating blend solutions (at 2500 rpm) onto clean micro-glass substrates. After that, thermal annealing for organic films was carried out using a digitally controlled hotplate at 100 °C for 10 min under nitrogen atmosphere in a glove box.

For solar cell fabrication, the ITO-glass substrates (~10 Ω/\Box) were sequentially cleaned in an ultrasonic bath using acetone (twice) and methanol (once), rinsed with deionized water, and finally dried in flowing nitrogen. To increase the work function of the ITO electrode and to improve the electrical connection between the ITO and the organic active layer, a layer of PEDOT:PSS was spin-coated (2500 rpm) onto clean ITO-glass substrate in air and dried using hotplate at 100 °C for 10 min under nitrogen atmosphere in glove box.

On top of the insoluble PEDOT:PSS layer, the P3HT:PCBM and P3HT:PCBM:C6 blend solutions were spin-coated (2500 rpm) and dried at 100 °C for 10 min under nitrogen atmosphere in glove box. An approximately 100-nm-thick Al electrode was thermally deposited onto active layer using a vacuum deposition system at a pressure of about 3×10^{-4} Pa through a shadow mask to obtain 25 identical cells on one device with an active area of $3 \times 3 \text{ mm}^2$. Then, the devices were thermally annealed in a nitrogen atmosphere at 140 °C for 4 min to increase the connection between Al electrode and active layer. The complete organic solar cells, with the configuration as shown in Fig. 2, were stored in the dark under suitable pressure until measurement.

Measurements

The optical absorption spectroscopy was carried out for P3HT: PCBM and P3HT: PCBM: C6 blend films with varying the C6 dye concentration using a JASCO V-570 ultraviolet/visible/near-infrared (UV/vis/NIR) spectrophotometer. The external quantum efficiency measurement was performed for ITO/PEDOT: PSS/P3HT: PCBM: C6/Al solar cells with varying the C6 dye concentration using a halogen lamp and a monochromator.



Results and discussion

Effect of C6 dye, as a small-molecule sensitizer, on optical absorption and ordering of the solar cell active layer

It is important to know that, the spectral range of the optical absorption compared to solar spectrum, absorption value and absorption wavelength are the factors that affect the total number of absorbed photons by solar cell active layer, where the limitation in the light absorption across the solar spectrum limits the photocurrent of the solar cells. The number of absorbed photons and, therefore, the generated photocurrent in a solar cell increase when the absorption range and absorption peak value increase for solar cell active layer. The number of absorbed photons in a solar cell active layer, also, increases when the active layer absorbs in the red or near infrared region of the solar spectrum, where there is the dense number of the visible light photons.

Fig. 3(a) shows optical absorption of pristine P3HT: PCBM and P3HT: PCBM: C6 blend films with varying C6 dye concentration. As shown in this figure, the P3HT in pristine P3HT: PCBM film absorbs light at the wavelength of 486 nm, while the PCBM absorbs around 316 nm. At the first sight, one can observe the higher contribution of the PCBM molecules to optical absorption of the P3HT: PCBM solar cell active layer. This is attributed to higher concentration of the PCBM molecules as 20 mg per 10 mg of the P3HT in the solar cell blend. By this way, we try to identify the role of PCBM at higher concentrations in light harvesting and, therefore, in photocurrent of the organic solar cells.

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Fig. 3. Optical absorbance of (a) pristine P3HT: PCBM and P3HT: PCBM: C6 blend films and (b) C6 dye fragments in P3HT: PCBM: C6 blend films with varying C6 concentration.

After adding C6 dye with different concentrations as 0.2, 0.5, 1 and 5 mg into P3HT:PCBM blend, the contribution of the dye to optical absorption appears as an absorption peak which overlaps with P3HT peak from the left side, as shown in Fig. 3(a). The resulted peaks from this overlapping gradually increase in their values, tend to occur at lower wavelength and take little different shape of the original P3HT peak accompanied with a little broadening in the wavelength range by increasing dye concentration in the P3HT:PCBM:C6 blends. Adding C6 dye with the amounts 0.2, 0.5, 1 and 5 mg into P3HT:PCBM blend encourages the overlapped absorption peaks of the P3HT:C6 binary to occur at 484, 480, 476 and 462 nm, respectively. This blue shift in the absorption peaks is due to absorption of the dye itself at lower wavelength, while the different shape of the absorption peak refers to the shape of the individual peak of pristine C6 dye, as observed in an our previous work [21]. Therefore, by increasing dye concentration in the P3HT:PCBM:C6 blend films, the light harvesting increases through the increase in the absorption peak value with a little broadening in the absorption wavelength range. This means that, the solar cell active layer absorbs higher amounts of the light photons.

From optical spectroscopy presented in Fig. 3(a) we can observe that, the C6 dye does not alternate vibronic structure of the P3HT molecules in the solar cell active layer. Unchanged vibronic structure of the P3HT after adding C6 dye is indicated form unchanged shoulders which appear in P3HT absorption spectra around 550 and 595 nm, as shown in Fig. 3(a). These shoulders are not fluctuated by adding C6 dye in the solar cell blend. The vibronic structure is generally explained in semiconducting polymers by a higher crystallization or ordering of intra-chain interactions [22]. Therefore, we can conclude that, the C6 dye does not affect on atoms interactions on the long of polymer backbone, and, consequently, C6 does not affect on crystallization or ordering (conjugation length) of P3HT in the solar cell active layer.

The absorption of C6 dye in P3HT: PCBM: C6 blend film can be individually extracted from the whole absorption profile of the blend film using a method done by Gebeyehu et al. [23]. In this method the P3HT: PCBM film (without dye) is put as a reference of the spectrophotometer, and the P3HT: PCBM: C6 films (with different dye concentrations) are put in the sample position of the spectrophotometer. Using this method, the resulted absorption spectra are shown in Fig. 3(b). It appears from this figure that, the absorption peak of pristine C6 dye with different concentrations occurs at the same wavelength around 462 nm. In addition, the shape of the C6 peak shown in Fig. 3(b) matches that of the main absorption peak in P3HT: PCBM: C6 blend film having 5 mg concentration of the dye, as shown in Fig. 3(a). This indicates that, the concentration with 5 mg of the C6 dye dominates the value, shape and wavelength of the main absorption peak in P3HT:PCBM:C6 blend films can approximately quantitatively be estimated by comparing the Figs. 3(a) and 3(b). Hence, one can observe that, the adding 1 mg of the C6 dye into P3HT:PCBM blend increases the main absorption peak value by 12%. Also, the peak value of the main absorption in P3HT:PCBM blend film is increased with 41% after adding 5 mg of the C6 dye.

The matching in the wavelength (462 nm) between absorption peaks of both pristine C6 dye, as shown in Fig. 3(b), and P3HT after adding 5 mg of C6 dye in the blend, as shown in Fig. 3(a), indicates that the C6 dye does not enhance or impair conjugation length or ordering of the P3HT polymer. Therefore, we can conclude that, the C6 dye inside P3HT:PCBM:C6 blend is phase separated from the other active layer components. The absorption spectrum of the P3HT:PCBM:C6 blend films shown in Fig. 3(a) represents a superposition of the individual components in the blend (i.e. no interaction of involved materials in the ground states). Otherwise and depending on their molecular structures and orderings, there are other dyes which can enhance conjugation length of the P3HT in the solar cell active layer, as observed in our previous work [24]. In the later case, the improvement in conjugation length of the P3HT is indicated as a red shift of the P3HT absorption peak in the optical spectroscopy, which represents an efficient tool for detecting the organization of the molecules in crystalline domains as stated by Vanlaeke et al. [25]. The observed improvement in light harvesting by incorporation of the C6 dye into P3HT:PCBM binary may introduce a positive impact on the photocurrent and performance of the corresponding solar cells provided that, other factors, such as excitons dissociation, charge carrier generation and charge carrier mobility, are sufficiently satisfactory for improvement of the corresponding solar cells.

Effect of C6 dye, as a small-molecule sensitizer, on external photocurrent quantum efficiency of the polymer solar cell

Using external quantum efficiency (EQE) spectra, we try to illustrate behavior of polymer solar cell through the incorporation of a small-molecule sensitizer in the solar cell active layer. The EQE is defined as the ratio of the number of charges extracted out of the device to the number of incident photons. The EQE spectrum is an efficient and appropriate tool for the active material(s) which is responsible for charge carrier generation in the solar cell active layer. Figure 4 shows the EQE spectra of the ITO/PEDOT:PSS/P3HT:PCBM/A1 and ITO/PEDOT:PSS/P3HT:PCBM:C6/A1 solar cells with varying C6 dye concentration in the solar cell active layer, through the wavelength range from 300 to 700 nm. Figure 4 depicts two important features. The first feature is the contribution of the C6 dye to photocurrent in P3HT:PCBM:C6 devices which occurs around 440 nm, where the C6 dye absorbs light around 462 nm, as shown in Fig. 3(b). The second feature shown in Fig. 4 is the contribution of the C6 after adding large amounts of the dye into blends, even though the P3HT represents the main sensitizer and donor-polymer in these solar cells. The contribution of the polymer to photocurrent in p3HT:PCBM:(0.2 mg)C6 soar cells appears as an eminent peak at the wavelength of 550 nm, where the P3HT has optical absorption peak at 486 nm, as shown in Fig. 3(a).



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Fig. 4. EQE as (a) real value and (b) normalized to peak value of the ITO/PEDOT:PSS/P3HT:PCBM:C6/Al devices with varying C6 concentration.

At the same wavelength (550 nm), the contribution of polymer to photocurrent in other solar cells appears as a hump (plateau) which declines with increasing C6 dye concentration from 0.5 to 5 mg.

The origin of the two features appeared in Fig. 4 can be discussed in the following. We can illustrate and determine the origin of contribution of the C6 dye, as a sensitizer, to photocurrent of polymer solar cell using the following:

- 1. The comparison shown in Fig. 5 between photocurrents of both P3HT: PCBM: C6 solar cell with varying C6 concentration and pristine C6: PCBM solar cell extracted from our previous work [21]. From Fig. 5 we can note that, by increasing C6 concentration in the P3HT: PCBM: C6 solar cell, the EQE spectra tend to take the same shape of the C6: PCBM solar cell spectrum. This means that, the P3HT: PCBM: C6 solar cell has the C6: PCBM combination among with P3HT: PCBM, as a main combination, in the solar cell blend.
- 2. The energy-level diagram of the P3HT: PCBM: C6 solar cell as shown in Fig. 1. From this diagram, we can note that, it is energetically suitable to form two individual cell combinations in the P3HT: PCBM: C6 blend. These combinations are P3HT: PCBM and C6: PCBM, which represent donor: acceptor combinations. These combinations can independently generate charge carriers in the same solar cell structure.
- 3. The results of absorption spectroscopy mentioned in the previous section. From theses results we found that, the C6 dye in the P3HT:PCBM:C6 blends was phase separated from the other active layer materials.

4. Depending on all these indications we verify that, the C6 dye, as a small-molecule sensitizer, can form an additional cell combination (additional charge carrier generation site) in the P3HT: PCBM: C6 blend through the combination with PCBM. The C6: PCBM combination can generate charge carriers among with P3HT: PCBM, as a main combination, in the solar cell blend. We may determine the origin of the second feature appeared in Fig. 4 by using Fig.

5. In this figure we can note that, by increasing C6 concentration, the P3HT:PCBM:C6 solar cell becomes like a solar cell which contains only C6:PCBM combination. This means that, the C6:PCBM combination has the prominent and main role for charge carrier generation in the P3HT:PCBM:C6 solar cell. The absence of the role of P3HT:PCBM as the main charge carrier generator in the solar cell active layer may be attributed to the presence of C6:PCBM combination that restricts transport of charge carriers generated by P3HT:PCBM combination. The restriction by C6:PCBM combination is attributed to low hole mobility of the C6 short-chain molecules [12,26]. Therefore, the transport of charge carriers from C6:PCBM into electrodes may be more appropriate than the transport of charge carriers from P3HT:PCBM into electrodes through C6:PCBM. The disability of the C6 in efficiently conveying the generated charge carriers in the blends reduces the photocurrent of the P3HT and the photocurrent of the whole solar cell spectrum, as shown in Fig. 4(a).



Fig. 5. EQE of C6:PCBM device (solid line) and P3HT:PCBM devices with varying C6 concentration.

From the results presented in this work, we can suggest an appropriate photocurrent generation mechanism in the P3HT:PCBM:C6 solar cell. The photocurrent of the P3HT:PCBM:C6 devices is induced by light absorption of both C6 and P3HT, as detected in Fig. 3(a), followed by creation of bound electron-hole pairs (excitons).

These excitons diffuse to interfaces where they can be dissociated into free carrier pairs depending on the strength of the local field which results from the decreasing in electron affinities and ionization potentials of C6 and P3HT in comparison with those of PCBM, as shown in Fig. 1. Therefore, the interfacial electric field drives charge separation, and the generated electrons are transported by PCBM to Al cathode, while the generated holes are transported by both C6 and P3HT towards the ITO anode. When C6 dye concentration is increased in the solar cell active layer, the C6 dye restricts transport of charge carriers generated by P3HT as a result of poor hole mobility of the C6 dye in the blends.

It is known that the PCBM contributes to a large extent in the photocurrent of the polymer:PCBM solar cells, as discussed by Hoppe et al. [27]. Indeed, the EQE spectra of the investigated solar cells in Fig. 4 show the contribution of the PCBM to photocurrent at a wavelength around 340 nm. The contribution of PCBM to photocurrent is attributed to pronounced light absorption at about 316 nm, as shown in Fig. 3(a), as a result of high PCBM concentration in the solar cell blend. In the P3HT:PCBM solar cell, the PCBM is considered the best electron acceptor so far. This is because: (i) ultrafast (~50 fs) photo-induced charge transfer occurs between donor polymer and PCBM, (ii) PCBM exhibits high electron mobility, which may reach to 1 cm2/Vs [28], measured by field effect transistors and (iii) PCBM shows a better phase segregation in the blend films [29]. Therefore, the increasing PCBM concentration in the P3HT:PCBM solar cell may enhance electron mobility into electrode.

Conclusions

For improving our knowledge about polymer solar cells, we had incorporated small-molecule sensitizer (C6 dye) with different concentrations into P3HT: PCBM solar cell having the composition 1:2 wt.%. We found from the present study that, the light harvesting of the P3HT:PCBM:C6 solar cell active layer increased with increasing C6 dye concentration, through the increase in absorption peak value with a little broadening in the absorption wavelength range. 5 mg of the C6 dye improved the peak value of the main absorption in the P3HT: PCBM: C6 blend film with 41%. From optical absorption spectroscopy we found that, the C6 dye in the P3HT: PCBM: C6 blend was phase separated from the other active layer materials. Therefore, the C6 dye, as a short-chain molecule, did not vary crystallization or ordering (conjugation length) of the long-chain polymer in the solar cell active layer. This was indicated from (1) unchanged vibronic structure of the P3HT after adding C6 dye and from (2) matching in the wavelength between absorption peaks of both pristine C6 and P3HT after adding C6 dye into blend (i.e. unchanged absorption of P3HT before and after adding C6 dye). From photocurrent spectroscopy we found that, the incorporation of the C6 dye, as a sensitizer, into P3HT: PCBM binary contributed to photocurrent and formed an additional charge carrier generation site through the C6: PCBM combination, which was found among with P3HT: PCBM combination in the same solar cell. In the same time, the C6 dye, as a short-chain molecule, restricted the transport of charge carriers generated by P3HT as a result of low hole mobility of the C6 short-chain molecules. Therefore, the EQE of the P3HT and, also, the overall EQE spectrum value decreased with increasing C6 dye content in the blends. Depending on the present study we can conclude that, the presence of a conjugated and highly ordered dye (small or long-chain molecule) as a sensitizer incorporated in P3HT: PCBM binary may improve solar cell performance through the contribution to charge carrier generation and through the contribution to charge carrier transport into electrodes.

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