



Comparative study of absorption and photoluminescent properties of organic solar cells based on P3HT:PCBM and P3HT:ITIC blends

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Abstract

We have studied the thin films of P3HT:PCBM and P3HT:ITIC blends as a prospective active layer for Organic Solar Cells by absorption, photoluminescence, and short-circuit current density–voltage characteristics. The potential estimation of structural changes in such photovoltaic devices is shown by employing the variations in absorption and photoluminescence spectra. The comparative analysis of key features of the absorption and photoluminescence spectra can be used as an indicator of their structural and photophysical properties.

1 Introduction

Organic solar cells (OSCs) are currently attracting the close attention of developers of renewable energy sources due to the key advantages of such photovoltaic devices as compared to their inorganic counterparts, such as the low cost of raw materials, easy manufacturing techniques, lightness and design flexibility, etc. [1]. But, the recent record high value of power conversion efficiency (PCE) of OSCs nearly around 15% is still significantly inferior to those of widely used silicon-based solar cells [2]. This situation stimulates systematic research aimed at finding new materials and structures that would improve the efficiency of various stages of the photovoltaic process in OSCs. Poly(3-hexylthiophene), P3HT, is one of the promising materials that demonstrated extremely important structural and photophysical properties achieving high PCE of solar energy [3]. This organic polymer with distinct donor properties may form a bulk heterojunction (BHJ) structure in a thin film from its blend with certain acceptor materials, where separated donor and

acceptor phases are characterized by nano-scale domains [4]. In such a medium, the excitons produced at absorption of sunlight quanta will efficiently dissociate to free charges at the boundary of the two phases. The key characteristics of P3HT ensuring the higher efficiency of this process are the self-organization of its molecules at high temperatures, and the formation of a laminar structure with crystal properties, when hole mobility of the medium is also high $10^{-2} \text{ cm}^2/\text{V} \cdot \text{s}$ [5, 6]. In addition, the molecular weight of such a donor polymer material, as well as some other similar ones, is crucial for the performance and morphology of OSCs based on them (see, for example, [7]).

Some fullerene derivatives like, phenyl-C61(C71)-butanoic acid methyl ester, PC₆₁BM (PC₇₁BM), may form a nano-scale BHJ structure upon blending with donor polymer materials and due to their pronounced acceptor properties and high electron mobility, they are considered as a prospective acceptor material for OSCs [8]. The above-mentioned properties of P3HT and PCBM allowed development of OSCs with PCE up to 5%, which has been a significant step in research and development on the consistent increase in the efficiency of OSCs [9]. The absorption bands of P3HT in the spectral region of 450–600 nm, and PCBM at shorter wavelengths together provide efficient absorption of sunlight at wavelengths < 600 nm. However, such a limited spectral region of absorption and low intensity of solar radiation in this short-wavelength part of the spectrum are the major physical limitation to the PCE of such OSCs [10].

To widen the action spectrum of OSCs, a number of narrow-band organic materials with the intensive sunlight

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absorption in the long-wavelength spectral region have been proposed [11, 12]. Replacing PCBM with IDTBR, a narrow-band acceptor material with strong absorption in the long-wavelength region at 550 nm 800 nm in [13] allowed increasing the PCE of such a OSC up to 6.4%, although it is still lower than its estimated value using the corresponding absorption spectra of the blends P3HT:PCBM and P3HT:IDTBR. This mismatch is most likely due to the low values of charge mobility and other photoelectric characteristics of IDTBR.

Another narrow-gap organic material with intense absorption in the same long-wavelength region is ITIC, [3,9-bis(2-methylene-(3-(1,1-dicyanomethylene)-indanone))-5,5,11,11-tetrakis(4-hexylphenyl)-dithieno[2,3-d:2,3-d']-s-indaceno[1,2-b:5,6-b]dithiophene], is characterized by strong absorption of the light at wavelengths 550–800 nm, where the energy density of solar radiation is close to its maximum, and the blend P3HT:ITIC absorbs much more solar energy than P3HT:PCBM. In addition, in the composition, where ITIC is an acceptor, and P3HT is a donor, the values of energy offset $\text{HOMO}_{\text{donor}} - \text{HOMO}_{\text{acceptor}}$ and $\text{LUMO}_{\text{donor}} - \text{LUMO}_{\text{acceptor}}$ are sufficient for driving the dissociation of excitons at the boundary of the two phases [8, 14]. However, to date, many attempts to achieve high PCE results in the OSC, with an active layer from P3HT:ITIC blend, have not been quite successful. Many structural and photoelectric properties of P3HT:ITIC thin films have been identified to limit the efficiency of such OSCs at 1% [15], although, blended with other polymeric donor materials with similar structural and photoelectric properties, ITIC demonstrated PCE above 10% [16]. Numerous studies of OSCs based on P3HT have shown unique opportunities of this polymer for structural changes enhancing of PCE and ranked it as a unique donor material. In addition to the energetic advantages of employing the P3HT:ITIC blend as an active layer of OSC, the possibility of identifying physical indicators does not lose its relevance that would allow controlling the photoelectric and photovoltaic characteristics of such device [17, 18]. Based on the above-mentioned arguments, a comparative study of the absorption and photoluminescence spectra of P3HT, PCBM, ITIC thin films, and their blends—P3HT:PCBM and P3HT:ITIC, grown under identical conditions, as well as their variations during thermic annealing was carried out in this work. The photovoltaic parameters of OSCs with active layer from these two blends, and the effect of thermal annealing on them were also studied. The physical aspects were also analysed which could have the aptitude to limit the PCE in such organic solar energy devices.

2 Materials and methods

P3HT was purchased from Sigma-Aldrich (USA, purity 99.995%), PCBM (PC₆₁BM or PC₇₁BM) were purchased from American Dye Sources (Canada, purity 99.5%), ITIC was purchased from Solarmer Materials Inc., PEDOT:PSS (Clevious PVP A14083) was bought from H.C.Starck. ITO glass substrates with a sheet resistance of 15 Ωsq^{-1} were purchased from Shenzhen Display (China). ITO glass substrates were first cleaned with detergent, and then sequentially cleaned with acetone, ultrapure water, and isopropyl alcohol in an ultrasonic bath for 13 min, respectively (see details in [19]). The ITO glass substrates were dried with N_2 and treated with O_2 plasma for 6 min to improve the wettability. PEDOT:PSS layer of 40 nm was fabricated on the ITO glass substrates by spin-coating at 4000 rpm for 20 s, and then, was thermally treated at 160 °C for 20 min. The solutions of P3HT, PCBM, ITIC and blend solutions of P3HT:PC₇₁BM and P3HT:ITIC (1:1 molar ratio) were dissolved in dichlorobenzene (DCB) with a concentration of 18 $\text{mg} \cdot \text{ml}^{-1}$. All the solutions were stirred overnight at room temperature for good solubility. Then, all the solutions were spin-coated at 800 rpm for 50 s ($\approx 100 \text{ nm}$) on the ITO/PEDOT:PSS surface. Then, a solution of 0.2 $\text{mg} \cdot \text{ml}^{-1}$ Poly[(9,9-bis(3-(*N,N*-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctyl fluorene)], PFN, in methanol was spin-coated onto the active layer, which has 5 nm of thickness. At the final stage, the substrates were pumped down to high vacuum ($5 \times 10^{-4} \text{ Pa}$), and 5 Al strips with a thickness of 100 nm and an area of 10 mm^2 each were thermally evaporated onto the active layer through shadow masks to define the active area of the devices. The current density voltage (*J*–*V*) characteristics were recorded with a Keithley 2420 source measurement unit under simulated 100 $\text{mW} \cdot \text{cm}^{-2}$ (AM 1.5 G) irradiation from a Newport solar simulator. In these measurements, the current density was determined by the total current through all five strips, which significantly reduced its divergence from sample to sample. In most of the experiments with P3HT:PCBM blends PC₇₁BM was used as an acceptor material because of its more intensive and wider spectral range of absorption [20].

Multilayered structures of glass/ITO/PEDOT:PSS/P3HT:PCBM/PFN/Al, glass/ITO/PEDOT:PSS/P3HT:ITIC/PFN/Al (in short, P3HT:PCBM and P3HT:ITIC structures, respectively) were, essentially, photovoltaic devices, in which, the major photovoltaic characteristics were also measured (see below). And, three other structures, glass/ITO/PEDOT:PSS/PCBM/PFN/Al, glass/ITO/PEDOT:PSS/ITIC/PFN/Al and glass/ITO/PEDOT:PSS/P3HT/PFN/Al (in short, PCBM, ITIC and P3HT structures, respectively), which differed from

the two above-mentioned only by the composition of the absorption layer, served as a comparative analogous for the study of variations in the absorption and photoluminescent characteristics induced by structural and energy changes upon mixing of the donor and acceptor materials and thermal annealing. Three samples were prepared from each solution under similar conditions and all the measurements were carried out on each of them and their average values were used in relevant calculations. The absorption spectra of these structures were measured in the region of 350–900 nm using a UV-1280 spectrophotometer (Shimadzu, Japan), and the photoluminescence spectra were measured in the region of 500–1000 nm at excitation with the light of 532 nm using a Renishaw 2000 spectrometer (UK). The thicknesses of the deposited films in the structures studied were measured using a DEKTAK 150 profilometer (Veeco, USA) with an accuracy of 0.6 nm.

3 Results and discussion

Absorption spectra Figure 1 shows the absorption spectra of PC₇₁BM, P3HT and ITIC structures of the same thickness, 100 nm. The light absorption in the substrate—glass/ITO/PEDOT:PSS, as well as in the PFN buffer layer was insignificant (< 0.05), in the all spectral regions of measurements. Therefore, the spectra shown are only referring to the absorption in PC₇₁BM, P3HT or ITIC films, respectively. Absorption of PC₇₁BM, as shown in the Fig. 1, decreases quite smoothly with increasing wavelength. In its spectrum along with the short-wavelength band at 380 nm there is a wide, but weak band in the region of 450–600 nm, depending on the aggregation state of the molecules [21]. P3HT of the same thickness has much stronger absorption, and its spectrum consists of three bands with peaks at 510, 550 and 600 nm, which are related to the main electronic transition

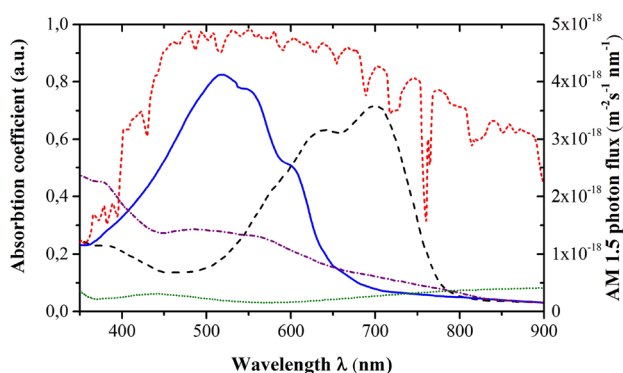


Fig. 1 The absorption spectra of the structures PC₇₁BM (dashed–dotted line), P3HT (solid line), ITIC (dashed line), substrate–glass/ITO/PEDOT:PSS (dotted line) and the spectrum of solar radiation on the Earth surface (short dashed line)

and its vibrational overtones [22]. The ratios of intensities of these bands characterize the degree of structural ordering of such polymer [23]. And, the absorption spectrum of ITIC consists of two long-wavelength bands with peaks at 650 nm and 700 nm, providing intensive absorption of sunlight at 400–775 nm in the blend P3HT:ITIC, while the absorption spectrum of the blend P3HT:PCBM is limited to wavelengths 350–625 nm.

In addition, the solar radiation in the spectral region of ITIC absorption is much higher than that in PC₇₁BM (the spectrum of solar radiation on the Earth's surface is also shown in Fig. 1). These two circumstances should ensure higher efficiency of the employing of the P3HT:ITIC blend as an active layer of OSC in comparison to P3HT:PCBM blend in terms of light energy utilization. According to the calculations described in [24], the contribution of PC₆₁BM to the PCE of OSC based on P3HT:PC₆₁BM has not exceeded 13%. From the experimentally measured spectral widths and absorption coefficients of the P3HT:PC₇₁BM and P3HT:ITIC structures and the energy spectrum of solar radiation known from literature, one can calculate the energy efficiency of sunlight utilization in these two cases [25]. Such calculations shown that in the similar conditions, the performance of OSC based on P3HT:ITIC should be three times higher than that based on P3HT:PC₇₁BM. However, the real values of the photovoltaic parameters, measured in the P3HT:ITIC structure, are happened to be more than ten times lower as compared to P3HT:PC₇₁BM structure (see below). Such an unexpected result was previously attributed to the low degree of crystallization of the P3HT polymer in the P3HT:ITIC film as compared to the P3HT:PCBM film, and poor separation of the donor and acceptor phases [14, 26]. It has to be noted that significantly higher PCEs were achieved in OSCs with some other polymeric materials as donor, and ITIC as an acceptor [27]. Thus, the structural properties and mutual complementary of these two components retains ordered arrangement of OSCs with optimal separation of donor and acceptor phases.

The most versatile method for obtaining active layer of OSCs with an ordered, crystalline structure and, at the same time, with the optimal separation of the donor and acceptor phases to form therein nano-scale domains is thermal annealing at higher temperatures (120–180 °C) or exposing to the vapor of appropriate solvents, to achieve potentially maximal PCEs [28]. To reveal the effect of structural changes in the active layer of OSC favoring the enhancement of PCE, and inducing relevant variations in the absorption and photoluminescence spectra of such device, we have studied absorption spectra of the P3HT:PC₇₁BM (Fig. 2a) and P3HT:ITIC (Fig. 2b) structures. The spectra of the freshly prepared samples were measured twice after their annealing at the temperature of 160 °C for 20 min (dashed and solid lines, respectively).

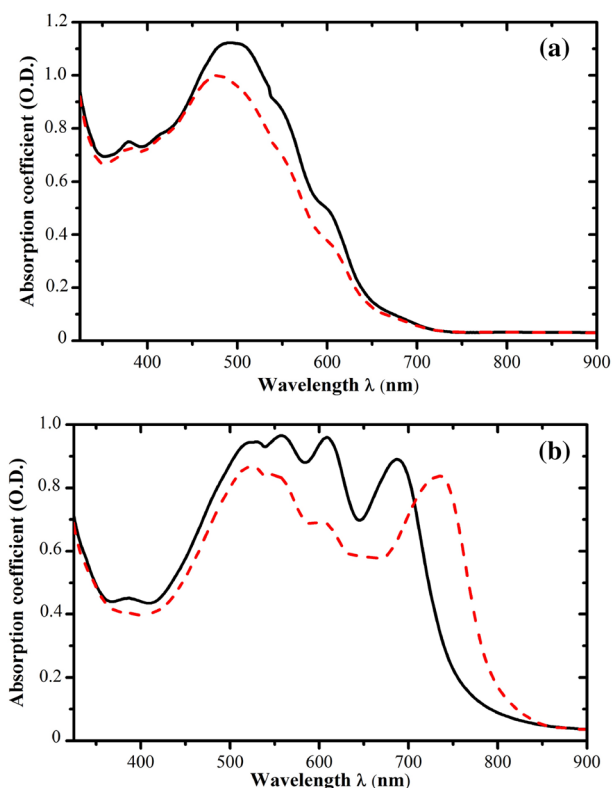


Fig. 2 The absorption spectra of the freshly prepared (dashed lines) and annealed at 160 °C for 20 min (solid lines) P3HT:PC₇₁BM (a) and P3HT:ITIC (b) structures

The peak of the absorption in freshly prepared P3HT:PC₇₁BM structure is shifted to the short wavelengths for 35 nm as compared to that of pure P3HT. However, after annealing, this peak undergoes a reverse shift towards the longer wavelengths for 25 nm. There is also a slight increase in the overall intensity of absorption after thermal annealing, especially, the bands associated to vibrational overtones with peaks at 550 nm and 600 nm [28]. There have been some detailed studies of the structural changes in such materials during thermal annealing using XRD, electron microscopy and optical spectroscopy [4, 29, 30], which demonstrated the possibility of an ordered lamellar structure formation with intro- and intermolecular links in the P3HT polymer. These structural changes were accompanied by variations in absorption spectra similar to the above. After annealing, a slight increase in the absorption was also observed in both pure PC₇₁BM, and P3HT:PC₇₁BM film in the short wavelength part of the spectrum, that was, apparently, due to the formation of domains of donor and acceptor phases. It is well known that the aggregation of PCBM molecules in organic solutions leads to the increased longer wavelength shift of its absorption bands [21].

The absorption spectrum of the freshly prepared P3HT:ITIC structure has an ITIC band with peak at 730 nm

along with the above-mentioned P3HT bands, i.e., the latter is shifted as compared to un-annealed ITIC peak for 30 nm (see dashed curves in Figs. 1 and 2b). Previously, such a long wavelength shift of the ITIC band has not been observed, even after thermal annealing or upon blending with polymer donor materials. In such conditions, spectral position of this band, does not change, or undergoes a small short wavelength shift due to the close location of the intensive absorption bands of donor polymers [31, 33]. Annealing of the P3HT:ITIC structure brought about similar changes, and the absorption bands of P3HT at 550 nm and 600 nm increased more intensively as compared to P3HT:PC₇₁B. The peak of ITIC absorption band underwent a short wavelength shift for 45 nm, and its spectral position in pure ITIC film, turned out to be at 700 nm. The XRD study of the P3HT:ITIC structure were carried out before and after thermal annealing [14], and it revealed that the freshly prepared sample of such blend has nearly amorphous structure. Annealing significantly increases its crystallinity, and is accompanied by various optical properties similar to the above. This situation suggests that the absorption spectra of the P3HT:PC₇₁B and P3HT:ITIC structure can be the indicator of characteristic structural changes in the active layers of corresponding OSCs. The measurement of the photovoltaic and photoluminescence characteristics of the BHJ structures studied confirmed this assumption (see below).

Photoluminescence spectra Photoluminescence spectra of the PC₇₁BM (a), ITIC (b) and P3HT (c) structures were obtained by exciting at the wavelength of 530 nm and the power of 5 mW (Fig. 3). The spectra are similar to the photoluminescence spectra of these substances in the solid-state form [28, 32, 34]. It should be noted that under the same measurement conditions and equal film thickness, the intensities of photoluminescence in such structures differed significantly from each other, which is caused not only by different values of their extinction coefficients, but also by different photoluminescence cross sections. As shown in Fig. 1, the absorption of P3HT film at 530 nm is three and four times higher than those of PC₇₁BM and ITIC, respectively, and PC₇₁BM molecules, having the form close to spherical, are characterized with extremely low photoluminescence quantum yield as compared to P3HT or ITIC [35].

The photoluminescence spectra of the freshly prepared P3HT:PC₇₁BM and P3HT:ITIC structures (solid curves in Fig. 4a, b, respectively), measured under the same conditions as their individual components, are similar in shape to the photoluminescence spectrum of P3HT. The low-intensity photoluminescence bands of PC₇₁BM at 710 nm and ITIC at 790 nm appear as barely noticeable shoulders of the broad and intense photoluminescence band of P3HT. However, the photoluminescence intensity in both P3HT:PC₇₁BM and P3HT:ITIC is significantly lower than that of pure P3HT film: whereas in the latter, the photoluminescence

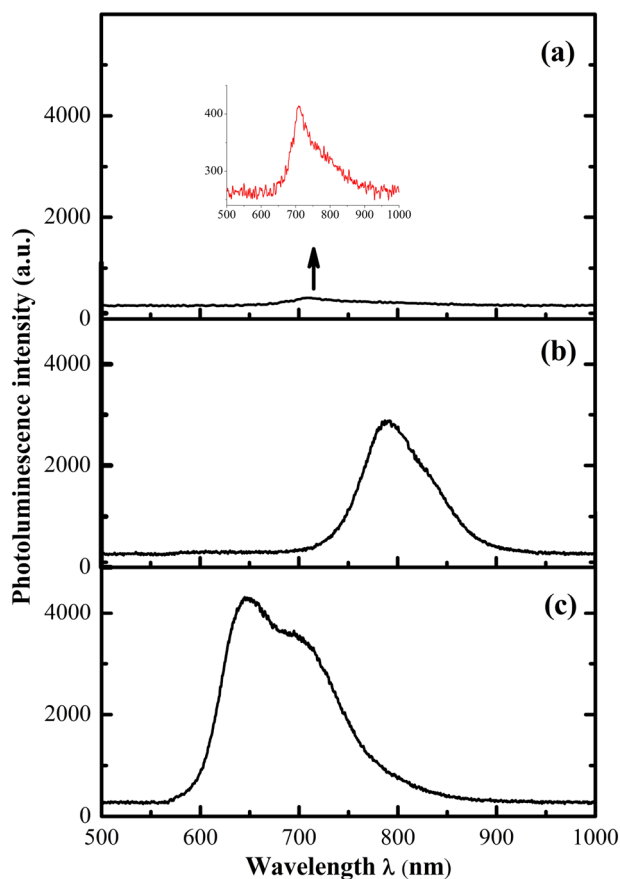


Fig. 3 The photoluminescence spectra of the PC₇₁BM (a), ITIC (b) and P3HT (c) structures measured in the same excitation conditions (530 nm, 5 mW). The inset shows the PC₇₁BM photoluminescence spectrum on an enlarged scale

intensity was 4400 units (Fig. 3c), in the P3HT:PC₇₁BM and P3HT:ITIC structures it was equal to 400 and 650 units, respectively (Fig. 4a, b). This result is due to fast and effective dissociation of the excitons to free charges at the boundary of the separated donor and acceptor phases in both structures [36].

The efficiency of exciton dissociation in OSC based on P3HT:PCBM active layer can be quantified by employing the quenching coefficient of the photoluminescence, determined by the formula [37]:

$$q = (I_{\text{P3HT}} - I_{\text{P3HT/PCBM}}) / I_{\text{P3HT}} \quad (1)$$

where, I_{P3HT} and $I_{\text{P3HT/PCBM}}$ —are experimentally determined photoluminescence intensities in P3HT and P3HT:PCBM films, respectively. The formula (1) has been used to calculate the photoluminescence quenching in OSC based on P3HT:ITIC. As per our measurements, the quenching coefficient of P3HT:PCBM structure was 0.91, which is in good agreement with literature data. Furthermore thin films of P3HT:PCBM blend with well-developed BHJ structure are

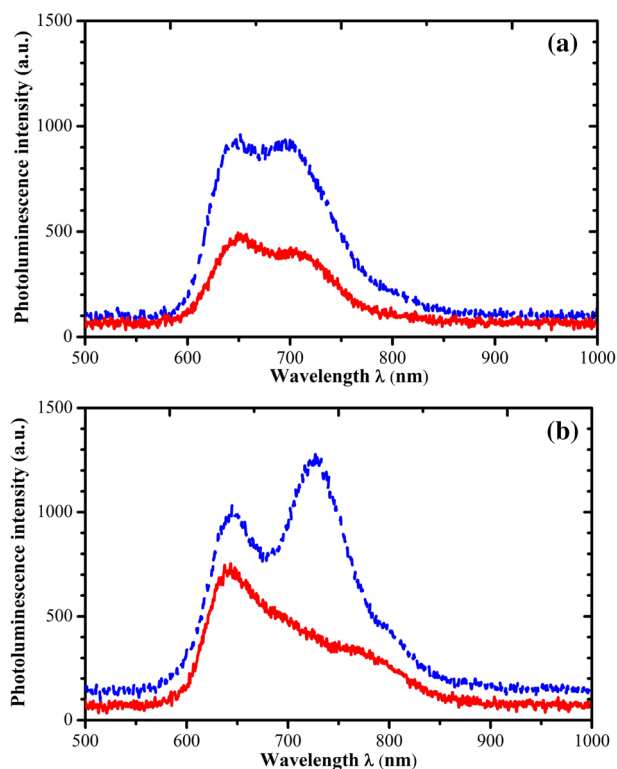


Fig. 4 The photoluminescence spectra of the freshly prepared (solid lines) and annealed (dashed lines) P3HT:PC₇₁BM (a) and P3HT:ITIC (b) structures

characterized with photoluminescence quenching coefficient > 0.9 under the optimal manufacturing conditions [36, 37]. And, quenching coefficient in the P3HT:ITIC structure, according to our measurements was < 85 . There are only few literature data on the photoluminescence quenching in P3HT:ITIC [15, 38], where the quenching coefficient of the P3HT:ITIC structure was very high, $> 95\%$, whereas, in the thin films of ITIC blend with other donor polymers, it was close to our data, 0.8–0.9, which might have a key importance for future development of highly efficient OSCs based on ITIC [33].

The annealing process of the P3HT:PC₇₁BM structure at 160° for 20 min caused a twofold increase in its photoluminescence. Such increase in photoluminescence of the analyzed structures after annealing could be explained by the well-developed separation of the donor and acceptor phases and the formation of their nano-scale domains, while the longer migration time and relevant dissociation of excitons at the boundary of these phases could increase the probability of their radiative decay. It should be noted, that the photoluminescence band at 700 nm, related to the vibrational overtone of the main electronic transition of P3HT, grew stronger than that of its band at 650 nm. This result is consistent with the above-described absorption data and the data from other reported works [28, 36].

The P3HT:ITIC structure annealed under the similar conditions demonstrated smaller, 1.3-fold increase in the short wavelength band at 640 nm, but there was more than a threefold increase in the long wavelength band at 730 nm. However, the structure of freshly prepared P3HT:ITIC displayed the intensity of the photoluminescence band at 640 nm, which is higher than that of freshly prepared P3HT:PC₇₁BM, while after annealing this band in P3HT:ITIC was noticeably higher than in P3HT:PC₇₁BM. This result can be interpreted as very low crystallinity of the freshly prepared P3HT:ITIC film, which increases faster than in P3HT:PC₇₁BM. Strengthening of the intro- and intermolecular links in the laminar structure of P3HT in the P3HT:ITIC thin film during annealing induces a sharp increase in the intensity of the photoluminescence band at 730 nm. It should also be noted that the stronger absorption of the freshly prepared P3HT:ITIC structure at wavelengths of 700–800 nm (see Fig. 2b) sharply decreases after thermal annealing, and enhances the observed effect.

Photovoltaic characteristics The main photovoltaic characteristics measured from the P3HT:PC₇₁BM and P3HT:ITIC structures before and after annealing at 160°C for 20 min are presented in Table 1 and Fig. 5.

As obvious from the table, the OSC based on the P3HT:PCBM structure demonstrated quite good results of the photovoltaic characteristics, with the exception of J_{sc} , which is somewhat less than its maximal values reported in the literature, apparently due to structural imperfections of the active layer in the measured samples [30]. A sharp increase in all photovoltaic characteristics occurred after

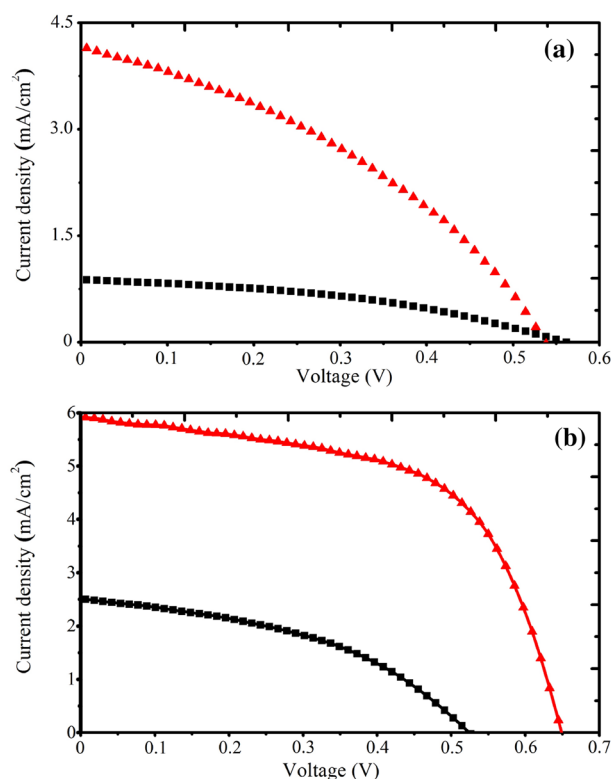


Fig. 5 The J–V curves of OSCs based on the P3HT:PCBM (triangles) and P3HT:ITIC (squares) structures before (a) and after (b) thermal annealing

Table 1 Photovoltaic characteristics of the OSCs based on P3HT:PCBM and P3HT:ITIC structures

Active layer	Open circuit voltage, V_{oc} (V)	Short-circuit current density, J_{sc} (mA/cm ²)	Fill factor (%)	Power conversion efficiency, PCE (%)
Un-annealed P3HT:PCBM averaged	0.536	4.15	37.2	0.83
No. 1	0.526	3.86	34.5	0.78
No. 2	0.521	3.94	37.9	0.74
No. 3	0.562	4.65	39.2	0.97
Annealed P3HT:PCBM averaged	0.648	5.92	58.5	2.25
No. 1	0.651	6.34	60.1	2.43
No. 2	0.646	5.74	58.2	2.18
No. 3	0.647	5.68	57.6	2.14
Un-annealed P3HT:IT averaged	0.56	0.88	40.7	0.2
No. 1	0.543	0.942	42.7	0.218
No. 2	0.588	0.853	37.9	0.19
No. 3	0.549	0.845	41.5	0.192
Annealed P3HT:ITIC averaged	0.53	2.5	43.1	0.57
No. 1	0.546	2.6	43.5	0.58
No. 2	0.525	2.43	41.8	0.55
No. 3	0.519	2.47	44.4	0.57

annealing, which is an inherent feature of the polymer-based OSCs. The analyzed OSC based on P3HT:ITIC structure has more complicated behaviors because the annealing caused a slight decrease in V_{oc} , but dramatically increased J_{sc} . However, the maximal value of the latter (2.5 mA/cm^2) is more than two times less in comparison to annealed P3HT:PCBM structure (Fig. 5). By considering the significantly wider absorption spectra of the P3HT:ITIC film as compared to P3HT:PCBM, these circumstances can be attributed to the strong disorder of the P3HT phase in such a blend [15].

4 Conclusions

P3HT as a donor polymer material with unique structural and photophysical properties demonstrated potential of constructing highly efficient OSCs in composition with a well-known acceptor material PCBM. However, in composition with the latest narrow-band acceptor material ITIC, despite the extremely favorable spectral complementarity of such a pair, P3HT is characterized to be of very low photovoltaic properties. A comparative study of the absorption, photoluminescence, and photovoltaic characteristics of the P3HT:PCBM and P3HT:ITIC structures revealed physical factors that could limit these features. And, the absorption and photoluminescence spectra of P3HT in such structures can be as an indicator of the degree of their structural order and the dissociation efficiency of excitons. For the study of relevant technological conditions and the manufacture of OSCs with the aim of increasing their PCE, the absorption and photoluminescence characteristics of such structures can be used as an informative indicator of their structural and photophysical properties.

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