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Impedance spectroscopy analysis of small molecule solution processed organic solar cell

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ABSTRACT

In this paper we study the transport-recombination mechanisms using impedance spectroscopy of organic solar cells (OSC) based on a blend of a small molecule, 7,7′-(4,4-bis(2ethylhexyl)-4H-silolo[3,2-b:4,5-b′] dithiophene-2,6-diyl)bis(6-fluoro-4-(5′-hexyl-[2,2′-bithiophen]-5-yl)benzo[c][1,2,5] thiadiazole) ($DTS(FBTTh_2)_2$) and 1-(3-methoxycarbonyl)-propyl-1-1-phenyl-(6,6) C70 ($PC_{70}BM$). We fabricate a cell with structure ITO/Poly(3,4-ethylenedioxythiophene)-poly(4-styrene sulfonate (PEDOT:PSS))/$DTS(FBTTh_2)_2$:$PC_{70}BM$/Ca/Al that exhibits $J_{sc}=10.2$ mA/cm^2, $V_{oc}=0.816$ V and FF=65% resulting in a PCE=5.4%. We model the impedance behavior using two circuital models, the parallel R-CPE and the transmission line model proposed by Belmonte et al. [1]. We compared the results to those obtained for OSC based on a standard poly(3-hexylthiophene) (P3HT): 1-(3-methoxycarbonyl)-propyl-1-1-phenyl-(6,6) C61 ($PC_{60}BM$) blend with structure ITO/PEDOT:PSS/P3HT:$PC_{60}BM$/LiF/Al. We find that in the case of the small molecule based OSC diffusion dominates over recombination for this thickness, $L=125$ nm, even at high frequencies. We calculate the effective carrier lifetimes and mobilities for both structures using both models. Average electron mobility calculated for the small molecule cell is around 4–6.4 $\times 10^{-3}$ cm^2/Vs, slightly higher than that obtained for the standard blend which is around 2×10^{-3} cm^2/Vs.

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1. Introduction

Bulk-heterojunction (BHJ) organic solar cells (OSC) have the potential to become real alternative to silicon-based devices due to the advantages they offer: lightness, thinness and potential flexibility [2,3]. Besides, the active layer material is solution processable, allowing low-cost fabrication techniques such as spin coating for small areas and different printing techniques in a continuous Roll-to-Roll process for large areas [4–6]. Although blends of conjugated polymers and fullerene derivatives have been one of the most popular due to the high efficiencies achieved [7] recently, devices based on blends of small molecules and fullerene derivatives have achieved high efficiencies, up to 9% [8]. As donor materials, small molecules offer additional advantages over polymers: the molecular structure is better defined and therefore the molecular weight and the material purity have low variations from batch to batch or among different materials supplier companies. Besides, in small molecules chemical properties can be finer tuned which also results in a better tuning of optical, electronic and physical characteristics.

Impedance Spectroscopy (IS) has been successfully used both in inorganic [9,10] and OSC to obtain valuable information about kinetics and energetic processes governing the device performance [11]. It is also a valuable tool to observe bulk and interfacial electrical properties that cannot be observed in direct current regime [12]. This technique consists in applying a small sinusoidal voltage superimposed on a bias voltage. By fitting the frequency response of the electrical impedance to a circuit model, information about cell resistances and capacitances can be extracted. In the literature different equivalent circuit models have been proposed to simulate the impedance of OSC: i) the simple parallel RC model, which results in perfect semicircular Cole–Cole diagrams, ii) the parallel R-CPE model used in [13–15], that results in depressed circular diagrams. This circuit includes an extra fitting parameter to model non ideal non-homogeneities such as porosities, roughness and surface states iii) The Mani parallel model [16] which consist in a parallel R_1CPE_1 in parallel with a series combination of R_2 and a second constant phase element, CPE_2 iv) Belmonte transmission line model, which includes distributed transport resistors, R_t, standing for carrier transport, and a distributed chemical capacitance, C_μ, in parallel with a recombination

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resistance, R_{rec} [1,17]. Impedance spectroscopy allows the determination of different electronic parameters, such as the built-in voltage, the doping concentration, the effective recombination lifetime and the diffusion time. From the diffusion time, diffusion coefficient and carrier mobility can be obtained [12,15,1]. Some authors have also used this technique to obtain the electron and hole carrier density and the density of states function [11].

In this paper we present a study of OSCs based on two different blends, the standard P3HT:$PC_{60}BM$ and a new blend that uses a small molecule as donor material, pDTS($FBTTh_2$)$_2$:$PC_{70}BM$. The layer structure is ITO/PEDOT:PSS/P3HT: $PC_{60}BM$/LiF/Al and ITO/PEDOT:PSS/pDTS($FBTTh_2$)$_2$:$PC_{70}BM$/Ca/Al, respectively. Cells have been optimized in terms of efficiency by adjusting some fabrication parameters such as ratio, concentration, annealing conditions, etc. Current density–voltage (J–V) curves are given and cell performance parameters are compared. Impedance spectra have been measured in dark at different bias voltages, and under varying illumination densities (at open circuit conditions, V_{OC}). Impedance data have been fitted with two models (the parallel RCPE and the transmission line model) and the extracted circuital parameters have been related to the transport processes occurring in the organic semiconductor.

2. Experimental details

2.1. OSC fabrication

Commercial ITO coated glass substrates (Präzisions Glas & Optik GmbH, 25 mm × 25 mm) were first manually washed in aqueous detergent and then sonicated for 10 min twice in acetone, isopropanol and de-ionised water. Substrates were blow dried with nitrogen and treated with UV ozone for 20 minutes.

PEDOT:PSS suspension (Clevios P VP Al4083) was filtered (with 0.45 mm PVDF filter), spin coated at 3.000 rpm for 60 s and annealed on a hot plate at 130 °C for 10 min yielding a thickness of 35 nm. The active layer blend of DTS($FBTTh_2$)$_2$:$PCB_{70}M$ (P3HT:$PC_{60}BM$) was spin coated and annealed on a hot plate at 150 °C yielding an active layer thickness of 125 nm (220 nm). Finally, a thin layer of 20 nm Ca (0.5 nm LiF) followed by a layer of Al (100 nm) were thermally evaporated on top of the device with a vacuum pressure lower than 10^{-5} mbar.

2.2. OSC characterization

J-V curves were measured using a Keithley 2400 Sourcemeter and a Steuernagel SolarCellTest 575 sun-simulator. Impedance spectra were measured using an IM6 Electrochemical Workstation from Zahner-elektrik. Samples were measured inside a flow cell, where a constant nitrogen flow was set in order to avoid degradation. A halogen lamp was used for illumination. Impedance spectra were recorded by applying a small voltage perturbation (20 mV rms) at frequencies from 1 MHz to 1 Hz in dark conditions (for voltages varying from −3to 1 V). Illumination measurements were performed varying the irradiation intensity up to one sun (i.e., simulated AM 1.5 G, 1000 W/m^2), under open-circuit conditions.

3. Results and discussion

Fig. 1 shows the device layer sequence together with the molecular structure and the energy levels schematic. LUMO and HOMO levels of p-DTS($FBTTh_2$)$_2$ have been reported by Poll et al. [18] using cyclic voltammetry and optical measurements. The difference between the HOMO level of donor molecule and

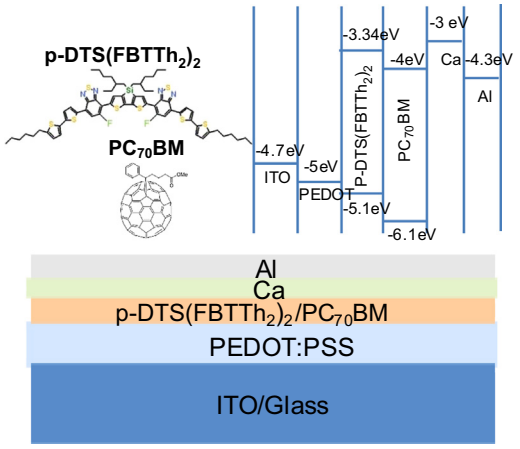

Fig. 1. Molecular structure, energy level diagram and device layer sequence.

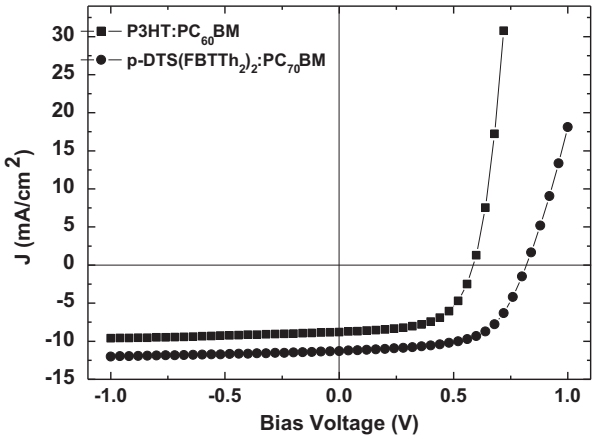

Fig. 2. J–V characteristic of the p-DTS($FBTTh_2$)$_2$:$PC_{70}BM$ and P3HT:$PC_{60}BM$ solar cells under simulated AM1.5G (100 mW/cm^2) irradiance.

the LUMO level of the acceptor (fullerene derivative) is related to the cell V_{oc} [19,20]. Previous results show that this photoactive blend p-DTS($FBTTh_2$)$_2$:$PC_{70}BM$ leads to high values of V_{oc} ranging between 0.77–0.81 V and thus resulting in an improved PCE of up to 7% [18,8].

Fig. 2 shows the J–V characteristic of the bulk heterojunction small molecule solar cell under simulated AM1.5G irradiation (100 mW/cm^2). This solar cell exhibits J_{sc}=10.2 mA/cm^2, V_{oc}=0.816 V and FF=65% resulting in a PCE=5.4%. For the sake of comparison, Fig. 2 also displays the J–V of a standard P3HT:$PC_{60}BM$ with similar layer structure both fabricated and measured under the same conditions. This standard cell exhibits J_{sc}=8.8 mA/cm^2, V_{oc}=0.574 V and FF=62%, leading to PCE=3.2%.

Fig. 3 shows the impedance spectra in dark conditions from 1 Hz up to 1 MHz for the small molecule solar cell at different bias voltages. Experimental data resemble the typical semicircle shape that can be accurately modeled with a simple parallel RC circuit. Solid lines show the fit. The capacitance C extracted from the fit is attributed to the depletion region capacitance due to the band bending at the contact [1]. This capacitance follows a voltage dependence according to the Mott–Schottky expression,

$$C^{-2} = \frac{2(V_{bi} - V)}{A^2 e \varepsilon \varepsilon_0 N_A} \quad (1)$$

where V_{bi} is the built-in potential, N_A is the acceptor impurity density, A is the area (=0.09 cm^2), V the applied voltage and ε and ε_0 are the dielectric constant and vacuum permittivity

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