



In-situ investigation of interfacial effects on charge accumulation and extraction in organic solar cells based on transient photocurrent studies

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ABSTRACT

In organic solar cells, the interfacial and bulk photovoltaic processes are typically coupled based on charge transport and accumulation. In this article, we demonstrated that the in situ transient photocurrent measurements can be a powerful approach to separately investigate the interfacial effects on interfacial and bulk photovoltaic process. Based on this method, the effects of interfacial dipoles on charge extraction, accumulation, and recombination are solely studied by comparing Ca and Al devices with standard architecture of ITO/PEDOT/P3HT:PCBM/cathode. We observe that stronger interfacial dipoles can significantly decrease the charge extraction time and consequently increase the charge extraction efficiency. More importantly, stronger interfacial dipoles can also decrease the charge accumulation within the bulk photovoltaic layer. Furthermore, our experimental results indicate that the bulk-accumulated charges can act as recombination centers under device-operating condition, resulting in the recombination loss in photogenerated carriers. Clearly, our studies of transient photocurrents elucidated the charge extraction, accumulation, and recombination in OSCs.

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1. Introduction

Organic solar cells (OSCs) have become a promising low-cost photovoltaic technology due to its merits of transparency, possible printing fabrication, and mechanical flexibilities [1–3]. In the last few years, the power conversion efficiencies (PCE) have largely improved exceeding 10% [4] due to the effort from materials synthesis [5–8] and device engineering [9–12]. Recent developments have shown that the organic/electrode interfacial properties are a critical component in improving photovoltaic perfor-

mance [13–16]. Specifically, the interfacial properties not only directly affect the charge collection efficiency [13–17] but also strongly change charge transporting process, such as charge accumulation and recombination [18–22]. Therefore, further understanding the complex photovoltaic processes related to interfacial properties is essentially important for device performance optimization.

In OSCs, the photogenerated carriers are either collected at the respective electrodes to generate external photocurrent or recombine within the bulk-heterojunction (BHJ) layer to cause a loss [19–22]. It is worth to emphasize that the interfacial charge extraction can affect bulk charge accumulation and recombination under device-operating condition. Specifically, the charge transport occurs through bulk and interfacial channels with series connection

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toward the generation of photocurrent. The bulk charge transport and interfacial charge extraction are mutually coupled in transporting photogenerated carriers. An effective interfacial extraction can lead to a less charge accumulation in a bulk photovoltaic film and consequently enhances the bulk charge transport. This can lead to a less recombination of bulk charge carriers. It should be pointed out that the dynamic effects of the interfacial charge extraction on bulk charge accumulation and recombination have not been extensively studied. In this work, we use transient technology to investigate the internally connected interfacial extraction and bulk accumulation and recombination based on conventional P3HT:PCBM solar cells. In general, transient measurements including photo-induced charge extraction by linearly increasing voltage (photo-CELIV) [23–26], transient photovoltage [27–32], and transient photocurrent [33–39] can be used to characterize interfacial properties, bulk charge trapping, and bulk recombination. In this report, we use transient photocurrent measurements to particularly investigate the influences of interfacial dipoles on surface charge extraction and bulk charge accumulation and recombination. The aluminum (Al) and calcium (Ca) cathodes with different work-function are employed to alter the interfacial dipoles [40–42]. It has been reported that the interfacial properties, such as interface gap state and chemical reaction, are different due to the difference in work-function between Ca and Al cathodes [40,43]. Ultimately, the interfacial dipole moment and energy level alignment can largely be changed when different electrodes are used.

2. Experimental section

2.1. Device preparation

The bulk heterojunction devices used in this study were based on blends containing poly(3-hexylthiophene) (P3HT) and 1-(3-methyloxycarbonyl)propyl-1-phenyl [6,6] C61 (PCBM). The high-workfunction Al and low-workfunction Ca cathodes are employed to alter the interfacial dipoles. The P3HT was purchased from Nanostructured Carbon and used as the electron donor. The PCBM purchased from Luminescence Technology was used as electron acceptor. The ITO glass substrates with a sheet resistance of $10 \Omega/\square$ were cleaned by ultrasonic cleaning in lotion, ethanol and deionized water successively, followed by dried in an oven for 2 h. A 40 nm thick layer of poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) (Baytron P Al4083) was deposited on to the oxygen plasma-treated substrates by spin-casting with 4000 rpm for 1 min, and then the PEDOT:PSS films were annealed at 150 °C for 30 min. P3HT and PCBM were co-dissolved in ortho-dichlorobenzene (ODCB) solution with a total solution concentration of 36 mg mL^{-1} and weight ratio of 1:0.8 (P3HT:PCBM). This solution was spin-coated at 1000 rpm to form 120 nm-thick blend films. The films were then transferred into a vacuum chamber for cathode deposition. The Al (100 nm) and Ca/Al (20 nm/100 nm, layer/layers configuration) electrodes were deposited under the vacuum of 2×10^{-6} Torr. The devices with Al as cath-

ode were post-annealed while the Ca devices were re-annealed at 150 °C for 10 min in nitrogen gas. The active area determined by the cross of ITO and cathode is 0.09 cm^2 in this study.

2.2. Device testing

The photocurrent–voltage characteristics were recorded by using Keithley 2400 source meter under illumination of AM 1.5G 100 mW cm^2 from Newport solar simulator calibrated to a silicon reference cell. For transient photocurrent measurement without background illumination, a high-brightness 590 nm LED (Intelligent LED Solutions, ILH-ON04-YELL-SC201) was used as the light source and a Hewlett Packard (HP) 8114A pulse generator was used as the power source for the LED. For the transient photocurrent measurement with background illumination, a white high-brightness LED (Intelligent LED Solutions, ILH-ON04-NUWH-SC201) was used for background illumination with Keithley 2400 as power supply. The purpose of employing background light is to explore the effects of trap on photocurrent dynamics. Therefore, the light intensity should be intensity-tunable. At the same time, the pulse intensity cannot be too strong to avoid charge accumulation within the device. A silicon solar cell (Newport 91150V) was used as the reference cell for light intensity calibration. Devices were connected in series with Agilent 33522A pulse/function generator operating in DC mode for voltage-dependent photocurrent measurements. The Tektronix DPO 4104 digitizing oscilloscope with input impedance of $1 \text{ M}\Omega$ recorded the transient photocurrent by measuring the voltage drop across an external 51Ω resistor in series with the voltage supply and the devices.

3. Results and discussion

In OSCs, the extraction of photogenerated charge carriers is either bulk-limited by the BHJ layer properties or interface-limited by the organic/electrode interfacial properties [19–22]. To distinguish these two limited processes, we examined the photocurrent decay dynamics in short-circuit current condition as function of pulse light intensity. Fig. 1a and b display the transient photocurrent responses under a pulse photoexcitation of 500 ns square wave at 590 nm. The insets indicate the experimental data for normalized current decay characteristics with fitted curves by using a single exponential decay mode. The curve fitting gives charge extraction time $\tau_{1/e}$, shown in Fig. 1d (the details can be seen in Supporting Information). The $\tau_{1/e}$ is defined as the time for the photocurrent to decay to $1/e$ of its peak value. The pulse intensity dependence of $\tau_{1/e}$ can be then used to estimate the extraction limiting processes in OSCs. We can see in Fig. 1d that the extraction time $\tau_{1/e}$ linearly increases with pulse intensity for Al device, while the device with Ca cathode exhibits weak intensity dependence of $\tau_{1/e}$ of $0.72 \pm 0.06 \mu\text{s}$. There are two different cases, namely bulk-limited regime and interface-limited regime, to determine the dependence of light intensity on extraction time $\tau_{1/e}$. In bulk-limited regime, the charge extraction process is mainly determined by

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