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Distinguishing limits on the fill factor in organic solar cells processed from different solvents: Charge recombination kinetics vs. charge extraction

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ABSTRACT

We use transient photovoltage (TPV) and transient photocurrent (TPC) techniques to understand charge recombination kinetics in a highly efficient organic solar cells (OSCs) system based on a blend of benzodithiophene terthiophene rhodanine (BTR) and [6,6]-phenyl C₇₁-butyric acid methylester (PC₇₁BM), processing different solvents. The OSCs fabricated from mixed solvents show the highest PCE of 10.2%, mainly due to an improved FF of 75.4% as compared to the devices from pure organic solvent, while J_{SC} and V_{OC} remain unchanged. Despite of difference in FF, non-geminate recombination is found to be the dominant photocurrent loss mechanism in all of the devices, regardless of processing solvents. Introducing a figure of merit, which is ratio between charge extraction rate and recombination rate by using the recombination kinetics data, we are able to correlate the competition between them to the fill factor of the devices.

1. Introduction

Solution-processable polymer and small molecules organic solar cells have become a highlighted research area in recent years [[1-4](#page--1-0)]. Compared to polymer organic solar cells (P-OSCs), small molecules organic solar cells (SM-OSCs) have attracted much attention since their well-defined structures, high chemical purity and better material reproducibility [\[5-8\]](#page--1-1). With the rapid progress in recent years, small molecule organic solar cells have achieved notably high power conversion efficiencies (PCEs) over 11% in single-junction devices [\[5,](#page--1-1)[9](#page--1-2)] mainly due to the development of novel materials and optimization in device processing. Ref. [\[10](#page--1-3),[11\]](#page--1-4) However, these results are still far away from the theoretical efficiency, [\[12](#page--1-5)] clearly indicating that there is still much room for improving the photovoltaic performance of organic solar cells by optimizing in device processing. Various device processing techniques have been reported in order to improve the overall performance of small molecules OSCs, including incorporation of additives, [\[13](#page--1-6)[,14](#page--1-7)] thermal annealing, [\[15](#page--1-8)[,16](#page--1-9)] solvent vapor annealing, [17–[19\]](#page--1-10) and use of mixed solvent.[\[20](#page--1-11)–24] While existing efforts by researchers were largely focused on understanding the influence of processing techniques on the nanoscale morphology and charge transport properties of active layer, there have been comparatively little studies dedicated to the charge recombination kinetics in a working device.[[25\]](#page--1-12) In a number of efficient OSCs systems, investigations have shown that the competition between charge recombination and charge extraction determines the

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shape of photocurrent output, thus have big impact on device performance characteristics, including short-circuit current density (J_{SC}) open-circuit voltage (V_{OC}) and fill factor (FF) [\[26](#page--1-13)].

Here in this study, we analyze a highly efficient OSCs system from a blend of benzodithiophene terthiophene rhodanine (BTR, see [Fig. 1a](#page-1-0) for the chemical structure) [\[27](#page--1-14)] and [6,6]-phenyl C₇₁-butyric acid methylester (PC_{71} BM), processing from pure chlorobenzene (CB) solution, pure chloroform (CF) solution and a mixture of CB and CF, respectively. The OSCs fabricated from mixed solvents show the highest PCE of 10.2%, mainly due to an improved FF of 75.4% as compared to the devices from pure CB and CF solvent, while J_{SC} and V_{OC} remain unchanged. The improvement in FF is discussed with respect to recombination kinetics and charge collection properties, both characteristic rates are accessible via transient measurements, such as transient photovoltage (TPV) and transient photocurrent (TPC) techniques. We find that non-geminate recombination is the dominant photocurrent loss mechanism in all of the devices regardless of processing solvents, as revealed by reconstructions of the J-V curves from the measured charge carrier decay rate and the voltage dependent charge carrier density in the devices. We further compare the charge extraction rate and recombination rate by using the recombination kinetics data, and correlate the competition between them to the fill factor of the devices.

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Fig. 1. (a) Chemical structures of BTR. (b). The typical J-V characteristics the device based on a mixture of CB and CF with a volume ratio of 1:1, tested under AM 1.5G (1000 W m⁻²) illumination in air.

2. Results and discussion

In [Fig. 1](#page-1-0)b, we show typical current density versus voltage characteristic $(J-V)$ for three sets of OSCs based on BTR: PC $_{71}$ BM (1:1) differing on the use of solvent (pure chlorobenzene, pure chloroform and a mixture of CB and CF with a volume ratio of 1:1), tested under 1000 W m−² air mass 1.5 global (AM 1.5G) illumination in air. The devices were prepared according to the literature procedures [[17](#page--1-10)]. The results are summarized in [Table 1](#page-1-1) for comparison. Note that the devices from mixed CF/CB solvent perform better than those from pure CF or CB solvent, exhibiting an improved efficiency of 10.2% while the two sets of control devices show PCE of 8.59% and 9.49%, respectively. To the best of our knowledge, this is among one of the highest efficiency for OSCs based on small molecule donor materials [\[5,](#page--1-1)[9](#page--1-2)]. It is apparent that the improvement is mainly due to an increase in FF (from ~65.8%/69.4%–∼75.4%), while J_{SC} and V_{OC} of the devices are left nearly unchanged.

Based on the device performance, the improvement in FF is likely indicative of enhanced charge transport and efficient sweep-out of charge carriers prior to recombination, although high-resolution transmission electron microscopy (TEM) can provide insight into phase separation in nanoscale and the nanofibrils interpenetrating networks (Figure S1). In order to determine charge carrier mobility in the active

Table 1 The device performance and charge carrier mobilities with different solvents.

Solvent	V_{OC} (V)	J_{SC} (mA) cm^{-2})	FF(%)	PCE $(\%)$	μ_e (cm ² $V^{-1}s^{-1}$	μ_h (cm ² $V^{-1}S^{-1}$
CF.	0.93	14.0	65.8	8.59	7.6×10^{-5} 1.1×10^{-3}	
$CF:CB = 1:1$	0.93	14.6	75.4	10.2	3.5×10^{-4} 2.8×10^{-3}	
CB	0.92	14.9	69.4	9.49	2.1×10^{-4} 1.4×10^{-3}	

layer, we measure the J-V characteristics of electron-only and hole-only device with device architecture of ITO/Al/BTR: PC_{71} BM/Al and ITO/ PEDOT:PSS/BTR: PC₇₁BM/Au, respectively, and analyze the results using a field-independent space charge limited current (SCLC) model [[28\]](#page--1-15). Figure S2 shows the J-V characteristics of the electron-only and hole-only devices as obtained in dark. The charge mobilities was deduced by fitting the J-V characteristics with the Mott-Gurney law given by $J = \frac{9}{8} \varepsilon_0 \varepsilon_r \mu \frac{V}{L}$ $\frac{9}{8}$ ε₀ ε_r $μ\frac{V^2}{L^3}$, where *J* is the current density, ε₀ is the permittivity of free space, ε_r is the relative permittivity of the active layer, μ is the carrier mobility, L is the film thickness of the active layer, and V is the effective voltage which is determined by subtracting the built-in voltage (V_{bi}) from the applied voltage ($V = V_{appl} - V_{bi}$). The extracted electron and hole mobility in film from mixed solvent are $3.5 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{s}^{-1}$ and $2.8 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{s}^{-1}$, respectively, both are higher than those of CF-based or CB-based devices ([Table 1](#page-1-1)). With the increased electron and hole mobilities, the sweep-out time of the charge carriers under different applied voltage is expected to decrease as well, thus may result in a reduction in the steady state charge carrier density in the active layer and a reduction in non-geminate recombination rate, as illuminated by our previous study [\[29](#page--1-16)].

Indeed, the more efficient sweep-out of charge carriers in the mixed solvent device can be supported by the dependence of photocurrent on electric field [[30\]](#page--1-17). [Fig. 2](#page-1-2) shows the measured photocurrent density (J_{ph}) versus effective voltage (V_{eff}) of each device, where $J_{ph} = J_L$ - J_D (J_L and J_D are the current density under illumination and in the dark, respectively), while $V_{\text{eff}} = V_0 - V_{\text{bi}}$ is the effective voltage representing the strength for charge extraction (V_0 is the compensation voltage at which net photocurrent $J_{ph} = 0$ and V_{bi} is the built-in voltage of the device). When the effective voltage V_{eff} exceeds 1.1 V, J_{ph} reaches its saturation value (J_{sat}) for all devices, suggesting nearly all photo-generated excitons are dissociated into free charge carriers, and most of which are collected at the electrodes, with little geminate or non-geminate recombination. Therefore, voltage-dependent ratio J_{ph}/J_{sat} , can be used to assess the exciton dissociation efficiency and charge collection efficiency in the device. For example, under short-circuit condition, the ratio J_{ph}/J_{sat} are 99.4%, 99.6% and 99.7% for the CF-based, CB-based and mixed-solvent-based-devices, respectively, implying efficient exciton dissociation and charge carriers in all of the three sets of devices. At maximum power point (MPP), J_{ph}/J_{sat} is 86.7% and 87.6%, for the

Fig. 2. Photocurrent density versus effective voltage characteristics of devices from different solvents under AM 1.5G illumination.

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