



# TFSA doped interlayer for efficient organic solar cells

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## ABSTRACT

Organic solar cells based on bis(trifluoromethanesulfonyl)amide (TFSA,  $[\text{CF}_3\text{SO}_2]_2\text{NH}$ ) doped poly[N-9'-hepta-decanyl-2,7-carbazole-alt-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole) (PCDTBT) were fabricated to investigate the effect of molecular doping. By replacing poly(3,4-ethylenedioxythiophene) (PEDOT): poly(styrenesulfonate) (PSS) with a thin TFSA layer, we have found more efficient charge injection at anode/active interface enhanced photovoltaic performance. The doping effect is confirmed by photoemission spectroscopy that the Fermi level of doped PCDTBT shifts downward to its HOMO level and results in higher carrier concentration. The reduced injection barrier also evidenced by impedance spectroscopy that the real impedance of the TFSA doped PCDTBT solar cell decreases more than 50%. Using the molecular doping approach, the overall power conversion efficiency (PCE) was largely increased from 4.70% to 5.98%. Our results suggest that TFSA functions not only as a surface doping molecule, but also as an anode interfacial layer to replace the conventional PEDOT:PSS.

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## 1. Introduction

Polymer solar cell is an emerging technology for renewable photovoltaic with the merits of easy processing, low cost, light weight, as well as the transparency and flexibility [1–3]. To date, the state-of-art organic solar cell has reached the power conversion efficiency (PCE) threshold of 10%, which has great competitiveness against other emerging solar cell technologies [4,5]. The major challenges hindering the advance of organic solar cells are the short exciton migration distance, narrow light absorption, and low stability [2,6–8]. Hence, plenty of efforts have

been devoted to improving the device performance, such as incorporating second or third solvents (1,8-diiodooctane (DIO) or chloroform (CF)) to optimize donor–acceptor morphology and thus minimizing the recombination rate [9–12], adding quantum dots (QDs) or nanoparticles to enhance the light absorption with a broader spectrum [13–16], or employing tandem structure to increase the open circuit voltage [17,18].

Besides the above issues, the carrier injection barrier at electrode/active layer interface, one of the most important figures of merit for organic materials also plays a significant role in determining the device performance. Molecular doping, one of the facile method to manipulate carrier density has been investigated for a long time and be considered as an effective approach to control the electrical property [19–22]. Recently, researchers found that the device performance and photoconductivity could be efficiently

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improved by doping polymers (Poly(3-hexylthiophene-2,5-diyl)(P3HT), Poly[2,6-(4,4-bis-(2-ethylhexyl)-4H-cyclopenta[2,1-b,3,4-b']dithiophene)-alt-4,7(2,1,3-benzothiadiazole)](PCPDTBT)) with widely used organic p-type dopant tetrafluoro-tetracyanoquinodimethan (F4-TCNQ). The resulting polymers had downward shifted the donor Fermi level and the increased carrier density [23,24]. Moreover, a hole-collecting favourable contact on P3HT diodes can be created by means of p-type doping strategy [25]. On the other hand, bis(trifluoromethanesulfonyl)amide (TFSA) has been reported as a valid p-type chemical dopant for carbon based materials (carbon nanotube, graphene) due to its strong electron withdrawing group [26–28]. Schottky junction solar cells fabricated with TFSA doped graphene have exhibited a PCE of 8.6% [29]. In addition, in polymer light emitting diodes, the work function of graphene electrode was tuned by TFSA doping for efficient hole injection [30].

In the present work, the effects of TFSA on the performance of solution processed bulk heterojunction organic photovoltaics (BHJ-OPV) are investigated. It is found that by utilizing a thin layer of TFSA between ITO and the active layer, the hole injection barrier at anode/polymer interface is reduced and the carrier injection becomes more efficient. Moreover, the TFSA incorporated device (without PEDOT:PSS) shows an improved PCE of 5.98% when compared with the traditional ITO/PEDOT:PSS/active layer/LiF/Al structure (PCE of 4.70%). Therefore, it is suggested that besides acting as a p-type doping molecule at the anode interface, TFSA also has the potential to serve as an alternative buffer layer to replace the commonly used PEDOT:PSS, which is always regarded as the cause for device degradation [31–34].

## 2. Experiments

In order to demonstrate the effect of TFSA, we fabricated a control device with configuration of ITO (180 nm)/PEDOT:PSS(40 nm)/PCDTBT:PC<sub>71</sub>BM (80 nm)/LiF (~1 nm)/Al (100 nm). PCDTBT was purchased from 1-material Chemscitech, Inc., and used as received. Firstly, a pre-patterned ITO (conductivity: 10–15  $\Omega$ /square) glass was cleaned by detergent, deionized water, acetone and isopropanol in sequence, followed by oxygen plasma treatment for 90 s. Secondly, for control sample, a thin layer of PEDOT:PSS was spun-cast onto ITO glass with a thickness of ca. 40 nm, and then annealed at 145 °C for 10 min in air. For comparison, device without PEDOT:PSS, TFSA (dissolved in nitromethane and stirred overnight) was spun coated onto ITO and formed a thin film of ~20 nm. Thereafter, the substrates were transferred to a N<sub>2</sub> filled glove box. PCDTBT and PC<sub>71</sub>BM with a mass ratio of 1:3 were dissolved in a mixed solution composed of 1,2 o-dichlorobenzene (o-DCB) and chloroform (CF) with volume ratio of 1:1, and then spun-cast atop the PEDOT:PSS (or TFSA) layer to form the active layer, giving rise to a thickness of ~80 nm. Following that, a 20-min soft annealing and a further annealing at 120 °C for 10 min were conducted. Finally, a thin LiF interfacial layer (~1 nm) and a 100 nm thick Al electrode were deposited sequentially by thermal evaporation. For each condition, there are at least 60 solar cells were measured and the figures were drawing from the average value.

The evaporator was BOC Edwards Auto 306 and the active layer area of the device was defined by a shadow mask of 2 mm  $\times$  6 mm. Electrical measurements were performed by a semiconductor characterization system (Keithley 236) at room temperature in air under the spectral output from solar simulator (Newport) using an AM 1.5G filter with a light power of 100 mW/cm<sup>2</sup>. The light intensity was precisely calibrated by a calibrated silicon solar cell. The morphologies of the PCDTBT and PC<sub>71</sub>BM blend thin films were characterized by atomic force microscopy (AFM) in tapping mode. The thickness of the active layer and evaporated layers were recorded with a thickness monitor (Sigma SQM-160), and also verified by AFM.

## 3. Results and discussion

Fig. 1(a) shows the chemical structure of materials that used in this study. Fig. 1(b) shows the solar cell configuration and Fig. 1(c) presents the energy band diagram in which PEDOT:PSS is replaced by TFSA. Once TFSA is in contact with PCDTBT, electrons will transfer from PCDTBT to TFSA due to the strong electron-accepting nature of TFSA. As electrons move to TFSA, the Fermi level of PCDTBT will shift downward to its HOMO level as indicated by the green arrow in Fig. 1(c). Such doping effect is confirmed by photoemission spectroscopy measurement that will be discussed later.

As an initial step, the thickness and concentration of TFSA were optimized, as shown in Figs. 1s and 2s. Firstly, the thicknesses of TFSA were varied from 5 to 25 nm and the concentration was fixed at 20 mM. With only 5 nm TFSA, the  $V_{oc}$  was increased to 0.87 V, when further increased to 10 nm, the  $V_{oc}$  reached 0.9 V and kept constant in the following thicker films. Overall, with 20 nm TFSA atop, the highest PCE was obtained. There are two possible reasons for the increased  $V_{oc}$  with thicker TFSA. Firstly, the different penetration depth of TFSA to the bulk active layer. At thin TFSA layer, like 5 nm, it is thick enough to cover the beneath ITO electrode, on the top side, after spin cast the active layer, there will be interpenetration of active layer into TFSA. At thin TFSA layer, the active layer could penetrate through the TFSA and may contact with ITO electrode to some extent, so there will be current leakage pathway there. For thicker TFSA, active layer could no longer penetrate through it, therefore the chance of leaking current was reduced and lead to stable  $V_{oc}$ . Secondly, more TFSA, 10 nm or thicker film compared to 5 nm, facilitate more complete doping of PCDTBT at the interface. Generally the energy difference between the HOMO of the donor and the LUMO of the acceptor (effective bandgap) set the upper limit of the  $V_{oc}$  in organic solar cell. However, the work-function of electrode has strong influence on the  $V_{oc}$  below that limit. As the effective bandgap in PCDTBT:PC<sub>71</sub>BM is around 1.4 eV, improving the WF in this material system still can enhance the  $V_{oc}$ . So in the rest of the study, the film thickness was fixed at 20 nm. For the concentration variation (5–25 mM), the cell with 20 mM TFSA doping showed the optimum PCE. Therefore, the film thickness and concentration were determined to be 20 nm

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