



All-in-one solar cell: Stable, light-soaking free, solution processed and efficient diketopyrrolopyrrole based small molecule inverted organic solar cells

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ARTICLE INFO

Article history:

Received 25 November 2015

Received in revised form

28 December 2015

Accepted 12 January 2016

Available online 16 February 2016

Keywords:

Small molecule

Diketopyrrolopyrrole

Inverted organic solar cell

Light soaking

Degradation

Charge transport layer

ABSTRACT

Organic solar cells (OSC) based on diketopyrrolopyrrole (DPP) small molecule have achieved relatively high efficiency (7%) in recent times. 2,5-di(2-ethylhexyl)-3,6-bis-(5'-n-hexyl-[2,2',5',2'']terthiophen-5-yl)-pyrrolo[3,4-c]pyrrole-1,4-dione (SMDPPEH) is the workhorse material for small molecule OSC as it has good solution-processing capability. However, there was no previous report on improving the device stability in inverted organic solar cells (IOSC) by using this material. Furthermore, the degradation and light-soaking behavior of this material in IOSC are also not well-addressed. In this work, we have fabricated a stable, light-soaking free, solution-processed and efficient SMDPPEH:PC₆₁BM based IOSC for the first time. Fluorosurfactant modified PEDOT:PSS and fluorinated TiO_x (F-TiO_x) transport layers were used to circumvent the inherent processability and light-soaking issues. An exclusive study on the device stability and light-soaking characteristics were also carried out for the first time. The final device provides the following merits: (i) comparable material stability to P3HT polymer, suggesting a potential for further development of DPP materials for high-efficiency devices; (ii) F-TiO_x can be used universally to fabricate a light-soaking free device with a wide range of photoactive materials from polymers to small molecules and; (iii) higher device efficiency compared to the non-inverted counterpart were obtained when the modified transport layers were used.

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1. Introduction

Solution processed organic solar cells (OSC) have made significant progress in recent times with device efficiency reaching 10% [1,2]. However, the device stability of OSCs still needs improvement if they have to be used in outdoor conditions. One way of improving the device stability is by employing the inverted architecture. In this architecture, the hole and electron transport layer positions are interchanged, resulting in the charge carriers being collected in the opposite direction across the device compared to its non-inverted counterpart. Inverted organic solar cells (IOSC) provides better stability compared to non-inverted devices through the following key modifications: (a) shifting of the hygroscopic PEDOT:PSS away from the ITO surface to prevent unintended etching and (b) by replacing the low work function

metal contact (Al) with a high work function metal (Ag) to prevent oxidation [3,4]. Impressive studies have been carried out on P3HT:PC₆₁BM based devices, to understand its degradation behavior under standard operating conditions [5–8].

Small molecule based photoactive materials can be a suitable candidate for the purpose of commercialization and large scale production. In fact, Heliatek GmbH announced a record 12% efficiency for its organic solar cells based on small molecules (oligomers) recently [9]. Relative to their polymer counterparts, small molecules hold several advantages: (i) uniform and defined molecular structures, reducing batch-to-batch variation [10,11]; (ii) more versatile and ease of modification of the chemical structure [12,13]; (iii) higher charge mobility [14] and (iv) generally higher open-circuit voltage [15,16]. Small molecule based organic solar cells (SMOSC) have been widely adopted in OSC as photoactive layer to achieve an efficiency of over 9% [16–19]. Diketopyrrolopyrrole (DPP) is a commonly utilized chromophore in organic electronics [10,20–22]. DPP is an attractive building block because of its facile synthetic modifications, i.e. substitution of solubilizing alkyl groups and aromatic groups for energy level

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tuning [15]. Due to the rationale of the conjugated structure, it has a greater tendency to crystallize, resulting in a high mobility. This property is desirable in OSC to enable better control of the morphology for optimal charge transport properties [23]. OSC with device efficiency of 7% was achieved using solution-processed DPP based donor small molecules [22].

Among the family of DPP molecules, 2,5-di(2-ethylhexyl)-3,6-bis-(5''-n-hexyl-[2,2',5',2'']terthiophen-5-yl)-pyrrolo[3,4-c]pyrrole-1,4-dione (SMDPPEH), is one material variation that has easy solution-processing capability [10]. The solubility of the material originates from the hexyl and ethyl-hexyl side chains at the thiophene terminals and DPP building block respectively. These alkyl side chains act as the solubilizing groups in the material, enabling decent solubility in organic solvents such as chloroform and chlorobenzene [10,24–26]. The terthiophene groups attached to the DPP backbone is crucial to make the molecule more planar and well-ordered, which results in high carrier mobility compared to non-thiophene substituents counterparts [24]. To date, typical device efficiency of 2.8% was reported when SMDPPEH:PC₆₁BM was utilized as a photoactive layer in the conventional device architecture [24,27]. Farahat et al. have reported a device efficiency of 4.5% by using (3-chloropropyl)trimethoxysilane as an additive to the photoactive blend SMDPPEH:PC₆₁BM [25]. However, there have been fewer reports of IOSC utilizing small molecules as photoactive layer. Few of which includes thermal evaporated SubPc:C₆₀ system [28,29] and p-DTS(FBTTh₂)₂:PC₇₁BM [30], without much information on the DPP based small molecule systems. Furthermore, the study of this small molecule material, which has a less complicated configuration, is essential to provide a deeper understanding on the behavior of the DPP functional group in inverted architecture.

Though inverted organic solar cells (IOSC) usually exhibits better device stability compared to the conventional OSC under ambient conditions, there are still two inherent issues: (i) wettability issue and (ii) light-soaking issue. Firstly, coating of PEDOT:PSS onto the photoactive layer remains to be a serious technical challenge involved in the fabrication of these devices. The hydrophobic nature of the underlying photoactive layer produces a high surface tension upon contact with the hydrophilic PEDOT:PSS, causing a serious wettability issue. Without the aid of additives, PEDOT:PSS can hardly be coated onto the photoactive layer, resulting in low device efficiency even though it may give stable device lifetime. As a result, additives are generally used to modify the wettability of PEDOT:PSS on photoactive layers [31–34]. However, these methods often cause deterioration of device efficiencies due to the requirement of additional UV–ozone treatment on the photoactive layer to enable proper film quality [35,36]. Secondly, a reversible light-soaking treatment is required when n-type oxide is used as a selective electron transport layer (ETL) in IOSC. Since this is a reversible effect [37–39], the device has to be activated repeatedly at each dark-light cycle (every morning if used in outdoor application) before it can function in full capacity. Hence, this effect should be addressed for improved device practicality. A typical device with TiO_x or ZnO as electron transport layer requires 10–15 min under AM1.5G to reach its maximum efficiency [40–42]. Impurity-modified metal oxides have also been used by various groups to address the light soaking issue [41,43]. Recently, we have reported the use of Dupont™ Capstone® FS-31 (CFS-31) modified PEDOT:PSS as HTL and fluorinated TiO_x (F-TiO_x) as ETL to overcome the wettability and light-soaking issues in P3HT:PC₆₁BM and PTB-7:PC₇₁BM based inverted organic solar cells [41,44,45]. We have identified that both transport layers were able to consistently provide its respective functions in polymeric photoactive layers but their performances in a small molecule system remains unclear. Thus, it

is important to evaluate the compatibility of these transport layers in a small molecule based IOSC.

In this work, SMDPPEH:PC₆₁BM based inverted organic solar cell was fabricated using CFS-31 modified PEDOT:PSS as hole transport layer and F-TiO_x as electron transport layer for the first time by overcoming the wettability and light-soaking issues. We have investigated the device stability and light-soaking characteristics of this material in inverted organic solar cell. Different sets of device were exposed to air in dark, light in N₂ environment, light in ambient condition and prolonged light-soaking treatment. It was found that the small molecule is relatively stable in all the three degradation conditions, and that the F-TiO_x electron transport layer significantly reduces the device light-soaking time.

2. Materials and methods

2.1. Materials

Chemical bath deposited F-TiO_x was prepared from ammonium hexafluorotitanate ((NH₄)₂TiF₆, 99.99%, Sigma Aldrich) and boric acid (H₃BO₃, ≥ 99.99%, Sigma Aldrich) as precursors. For sol gel-TiO_x, Titanium isopropoxide (TTIP, 97%, Sigma Aldrich), acetylacetone (AA, Sigma Aldrich) and isopropanol (IPA, reagent grade, Aik Moh Paints & Chemical Pte Ltd.) were used. Regioregular poly(3-hexylthiophene-2,5-diyl) (P3HT, > 98%, Sigma Aldrich), poly({4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl}{3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophene-diyl}) (PTB-7, 1-Material) and 2,5-di(2-ethylhexyl)-3,6-bis-(5''-n-hexyl-[2,2',5',2'']terthiophen-5-yl)-pyrrolo[3,4-c]pyrrole-1,4-dione (SMDPPEH, 1-Material) were used as donor materials. Phenyl-C₆₁-butyric acid methyl ester (PC₆₁BM, Nano-C) and phenyl-C₇₁-butyric acid methyl ester (PC₇₁BM, Nano-C) were used as acceptor materials. Dichlorobenzene (DCB, Sigma Aldrich) and chlorobenzene (CB, Sigma Aldrich) were used as the solvent for donors and acceptors. 1,8-diiodooctane (DIO, Sigma Aldrich) was used as a solvent additive for PTB-7:PC₇₁BM. Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS, Clevios P VP Al4083, Ossila) were used as hole transport layer. Capstone® FS-31 (Dupont™) was added into PEDOT:PSS prior to spin coating. Silver (Ag) metal was purchased from K.J. Kurt Lesker & Co. P3HT:PC₆₁BM and PTB-7:PC₇₁BM IOSC were fabricated as control devices for the degradation study and the light-soaking study. All the above mentioned materials were used as received.

2.2. Solution preparation

The precursor solution for F-TiO_x films were prepared by mixing appropriate concentration of ammonium hexafluorotitanate ((NH₄)₂TiF₆, 99.99%, Aldrich) and boric acid (H₃BO₃). Both solutions were stirred separately for at least 10 min at room temperature before mixing. The stirred solutions were mixed and placed into a pre-heated bath with temperature at 40 °C before immersing ITO substrates into the bath for F-TiO_x deposition. The deposited film was finally annealed at 180 °C for 1 h. The precursor solution for sol-gel TiO_x was prepared by mixing titanium isopropoxide, acetylacetone, iso-propanol in volume ratio of 1:0.5:10.

P3HT:PC₆₁BM blend was prepared in the ratio of 1:0.8 (15 mg/ml of P3HT, 12 mg/ml of PC₆₁BM) in DCB. PTB-7:PC₇₁BM blend was prepared in the ratio of 1:1.5 (12.5 mg/ml of PTB-7, 18.75 mg/ml of PC₇₁BM) in CB (96%) and DIO (4%). SMDPPEH:PC₆₁BM blend was prepared in the ratio of 1:1 (10 mg/ml of both SMDPPEH and PC₆₁BM) in CB. All mixed solution were stirred and heated at 60 °C overnight in a N₂ filled glove box prior to spin coating.

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