



Alternative benzodithiophene (BDT) based polymeric hole transport layer for efficient perovskite solar cells



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ABSTRACT

Organic-inorganic perovskite cell has shown a great deal of interest in past few years due to its ability to achieve high power conversion efficiency (PCE). Use of charge transport layers such as n-type TiO₂ and p-type doped spiroOMeTAD in a n-i-p device architecture has shown enhanced perovskite solar cell device performance. Use of doped spiroOMeTAD as hole transport layer in the n-i-p device structure has been effective but possess disadvantages such as complex processing, use of corrosive additives, processing in ambient air for efficient hole doping. Here we report the study of solution processed benzodithiophene based polymer PBDTT-FTTE as an alternative hole transport layer to doped small molecule spiroOMeTAD. PBDTT-FTTE doped with 3% DIO (diodooctane) achieved PCE of 11.6% which was comparable to matching PCE of 11.6% obtained from using spiroOMeTAD as hole transport layer. We showed that unlike spiroOMeTAD, polymer PBDTT-FTTE is processed inside N₂ filled glove box and is easier to process as compared to spiroOMeTAD which requires processing in ambient humid air and is doped with additives mixed in corrosive solvent, causing degradation to perovskite layer underneath.

1. Introduction

Hybrid organic-inorganic perovskites have ABX₃ type chemical composition, where A is an organic or inorganic cation, B is a divalent metal cation, and X is a halide anion. Lead-halide based perovskite namely methylammonium lead triiodide (CH₃NH₃PbI₃) has been widely investigated in recent years because of its unique properties that are highly suitable for photovoltaic applications [1–5], compared to other low cost solar cell counterparts based on polymer [6,7], DSSC [8–11] and CZTS thin films [12]. Organic-inorganic perovskites have numerous advantages as they are solution processable with tunable band gaps, and superior charge-transfer properties which make them suitable for solar cell fabrication. Semiconducting perovskite (CH₃NH₃PbI₃) light absorber has been used to fabricate efficient solar cell in both planar p-i-n [13–16] and mesoporous n-i-p device [17,18] architectures. The first use of perovskites in solar cell application was reported by Miyasaka in 2009 where he used mesoporous n-i-p device architecture which exhibited photo-conversion efficiency (PCE) of 4%

[19]. Since then, there has been a tremendous progress in perovskite solar cells resulting in a PCE exceeding 20% [20]. Mesoporous n-i-p device structure has been most widely adopted architecture wherein popular doped small molecule 2,2',7,7'-tetrakis(N,N-di-p-methoxyphenylamine)–9,9-spirofluorene (spiroOMeTAD) has been used as hole transport material (HTM) [21,22]. Spiro-OMeTAD in its pristine form exhibits poor hole mobility and electrical conductivity leading to poor hole transport properties. Therefore, spiroOMeTAD is doped with p-type dopants like lithium bis(trifluoromethylsulfonyl)imide (Li-TFSI) along with 4-*tert*-butylpyridine (TBP) in order to improve hole mobility and electrical conductivity, making the processing complex and costly [23]. The dopants used in spiroOMeTAD is corrosive to perovskite film, and processing of spiroOMeTAD requires spin coating in ambient atmosphere, which allows exposing moisture sensitive perovskite films to humidity, causing structural degradation of perovskite. Therefore, there is a need for a simple solution processable hole transport layer having excellent electrical properties in order to fabricate stable, cost effective and efficient solar cells. An alternative HTM should be solution

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processable in inert atmosphere with least use of additives. Several organic materials have been reported as hole transport material as an alternative to doped spiroOMeTAD in perovskite solar cells and have shown similar or enhanced device performance. These materials include polymers [18,24,25], small molecules [26–28] and inorganic materials [27]. Semiconducting p-type donor polymer-acceptor (D-A) have been used as active layer in bulk-heterojunction solar cells [29,30] and also as HTMs in perovskite ($\text{CH}_3\text{NH}_3\text{PbI}_3$) based solar cells.

In this work, we report the use of a novel solution processed semiconducting p-type polymer Poly[4,8-bis(5-(2-ethylhexyl) thiophen-2-yl)benzo[1,2-b;4,5-b']dithiophene-2,6-diyl-*alt*-(4-(2-ethylhexyl) – 3-fluorothiopheno[3,4-b]thiophene-) – 2-carboxylate-2-6-diyl)] (PBDTT-FTTE) as hole transport layer that replaces spiroOMeTAD in organic-inorganic perovskite ($\text{CH}_3\text{NH}_3\text{PbI}_3$) solar cells. PBDTT-FTTE has been used in bulk heterojunction solar cells as donor and fullerene derivative as acceptor, leading to a PCE of 9.35% [31]. The polymer PBDTT-FTTE incorporates 2-ethylhexyl-thienyl group in benzodithiophene (BDT) unit which improves coplanarity of the main chain, thus enhancing charge transport properties of polymer. Further, PBDTT-FTTE is solution processable with no requirement of post-deposition thermal annealing. The highest occupied molecular orbital (HOMO) energy for PBDTT-FTTE lies ~ 5.2 eV, which makes the hole extraction highly efficient from perovskite film having valence band at ~ 5.4 eV. The use of polymer PBDTT-FTTE eliminates problems which are encountered when using spiroOMeTAD such as 1) perovskite film is protected from atmospheric humidity as polymer PBDTT-FTTE does not require ambient processing and (2) simple processing with no or least use of additives. These unique properties of polymer PBDTT-FTTE makes it suitable alternative to spiroOMeTAD.

To our best knowledge, there have been no report on use of BDT based polymer named PBDTT-FTTE as hole transporting material in perovskite solar cell. Herein, we demonstrate processing of PBDTT-FTTE polymer with and without additive (3% diiodooctane) as hole transport layer in perovskite solar cells and compare its photovoltaic performance with doped spiroOMeTAD based perovskite solar cell. We show that use of polymer PBDTT-FTTE with 3% DIO with simple solution processing in inert atmosphere exhibited similar photovoltaic performance as obtained with spiroOMeTAD based perovskite solar cells.

2. Experimental procedure

2.1. Materials

Methylammonium iodide ($\text{CH}_3\text{NH}_3\text{I}$) and 20 nm mesoporous TiO_2 (18NRT) were purchased from Dyesol. PbI_2 (99%) was obtained from Acros organics. Spiro-OMeTAD was purchased from Lumtec, Taiwan. PBDTT-FTTE polymer was purchased from Cal-OS Inc. All materials, were used as received.

2.2. Solar cell fabrication

Mesoporous n-i-p device architecture was used for fabrication as shown in Fig. 1a. Fig. 1b shows energy level band diagram of n-i-p solar cell using polymer PBDTT-FTTE as hole transport layer. Fluorine doped tin dioxide (FTO) coated glass substrates were cleaned by ultrasonication using soap water, distilled water, acetone and isopropanol for 20 min each. The substrates were dried with compressed nitrogen and plasma treated in presence of oxygen for 20 min. Anhydrous ethanol was used to make diluted solution of 0.15 M and 0.3 M from Titanium diisopropoxide bis (acetylacetonate) (75 wt% in isopropanol). Compact TiO_2 layer (0.15 M) was spin coated on top of FTO substrates followed by annealing at 200 °C for 10 min. Then 0.3 M compact TiO_2 layer was spin coated followed by annealing at 450 °C for 30 min. The mesoporous TiO_2 layer composed of 20 nm particle size (diluted in ethanol in 2:7 wt ratio) was spin coated at 3000 rpm for 40 s, followed

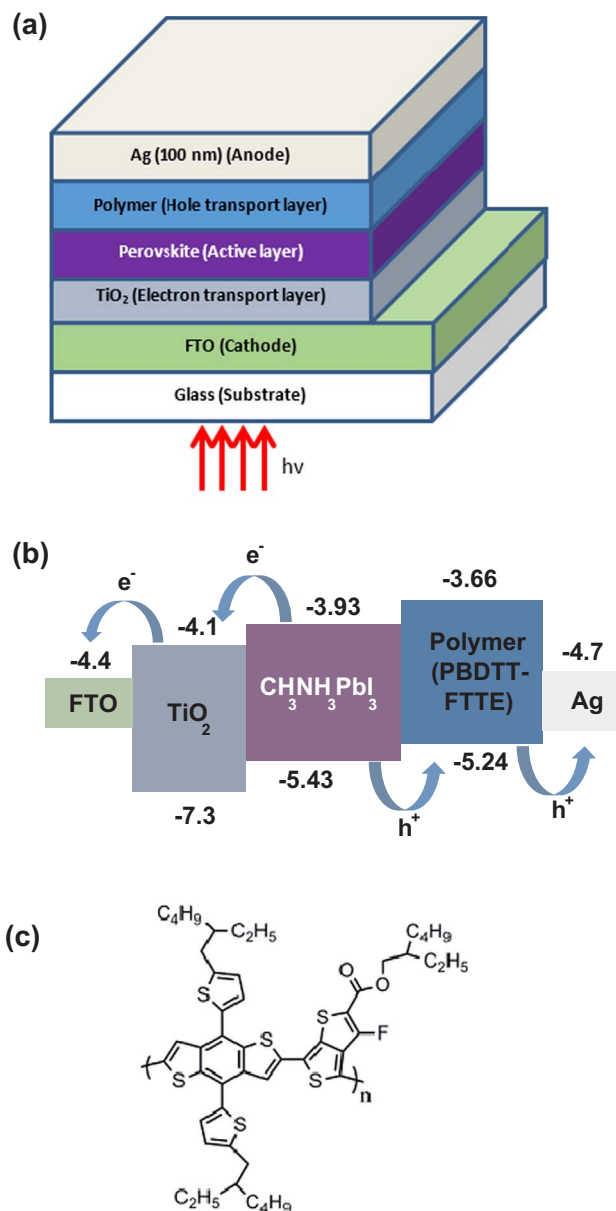


Fig. 1. (a) Device structure and (b) energy band diagram of the n-i-p solar cell. (c) Chemical structure of the polymer PBDTT-FTTE.

by annealing at 450 °C for 30 min. The mesoporous TiO_2 layer was treated with 25 mM TiCl_4 aqueous solution by immersion in the solution for 30 min at 70 °C followed by rinsing the films by de-ionized water and ethanol. The films were then annealed at 450 °C for 30 min. TiCl_4 treatment of TiO_2 layer was carried out to passivate the defects in TiO_2 and improve the interface between TiO_2 and perovskite, resulting in reduction of charge recombination at the interface [32,33]. The TiCl_4 treated TiO_2 films were then transferred into a nitrogen filled glove box for perovskite layer deposition. Two-step sequential dip coating method was used to deposit organic-inorganic perovskite layer. Lead iodide was dissolved in N, N- dimethylformamide (1 M) with stirring at 70 °C. PbI_2 precursor was spin coated at 4000 rpm for 40 s followed by annealing at 70 °C for 30 min. The annealed PbI_2 films were then dipped in 10 mg/ml of $\text{CH}_3\text{NH}_3\text{I}$ solution in 2-propanol for 60 s. The dip coated perovskite films were immediately dried by spin coating at 6000 rpm for 10 s, followed by annealing at 100 °C for 15 min on a hot plate inside a N_2 filled glove box. Spiro-OMeTAD solution was prepared by mixing 72.3 mg of spiroOMeTAD, 17.5 μL of 520 mg/ml lithium bis (trifluoromethylsulfonyl)imide salt in acetonitrile, and 28.8 μL of TBP

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