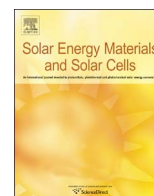




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Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat

Highly-efficient polymer solar cells realized by tailoring conjugated skeleton of alcohol-soluble conjugated electrolytes

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ARTICLE INFO

Article history:

Received 4 May 2016

Received in revised form

28 June 2016

Accepted 26 July 2016

Keywords:

Polymer solar cells

Small molecule conjugated electrolytes

Cathode interfacial modification

 π -conjugated skeletons

ABSTRACT

Compared with the popular conjugated polyelectrolytes as interfacial materials in polymer solar cells (PSCs), small molecule conjugated electrolytes (SMCEs) interlayers remains limited and the relationship between structures of small molecule electrolytes and device performance is still unclear. Herein, a series of alcohol-soluble small molecule conjugated electrolytes, combined with different conjugated skeletons and identical ammonium salt polar side chains, namely, **2T-(FN-Br)**, **3T-(FN-Br)₂** and **4T-(FN-Br)₂** were designed and synthesized as interfacial materials for the inverted polymer solar cells (I-PSCs), to explore the relevance between the π -conjugated skeleton and the device performance. As the conjugated bridge length and terminal group increased from **2T-(FN-Br)**, **3T-(FN-Br)₂** to **4T-(FN-Br)₂**, the photovoltaic performances were gradually improved. The improved device performance caused by the enlarged π -conjugated skeleton mainly resulted from the optimized interfacial contact, effective modulation on the morphology of the active layer and the improvement of charge mobility. Encouragingly, devices with **4T-(FN-Br)₂** as interfacial material achieved a high power conversion efficiency of 8.40%, based on thieno [3,4-b]-thiophene/benzodithiophene (PTB7): [6,6]-phenyl-C71-butyric acid methyl ester (PC₇₁BM) as light-harvesting layer. This work provided a new avenue to exploit novel promising interfacial modification materials by simply and slightly modulating the π -conjugated skeleton without complex molecular design.

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1. Introduction

Polymer solar cells (PSCs) with a blend of conjugated polymers and fullerene derivations as photoactive layer draw an enormous amount of attention, owing to provide a bright avenue to directly convert the terrestrial solar radiation into electricity by fabricating large-scale printing, low-cost, flexible devices [1–5]. Recently, great progress has been made in improving the power conversion efficiency (PCE) of PSCs which exceeded 10% for single-junction organic solar cells [6–8]. It has been proved that the conventional device lifetime is degraded by the use of easily-oxidized low work function metal cathode and the acid hole-transportation layer material poly(3,4-ethylenedioxyethiophene): poly(styrenesulphonic acid) (PEDOT: PSS) which is corrosive to the indium tin oxide (ITO) anode [5,9–11]. This accelerates the foundation of the inverted device in which the high work-function of ITO is applied as cathode electrode

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to improve the stability of device. However, energy loss stemming from the accumulation and exciton recombination still exist in the inverted devices due to the energy mismatch between the high work function of ITO (~ 4.7 eV) and the lowest unoccupied molecular orbital (LUMO = ~ 4.2 eV) of the fullerene acceptor [12–14]. Therefore, the key issue in the inverted polymer solar cells (I-PSCs) is the development of useful interlayers to tune the work function of the ITO to work as the suitable cathode [15,16].

In recent years, the novel organic conjugated electrolytic materials which are combined with polar groups on the side chains are very popular to develop as interfacial materials. Conjugated electrolytic materials included polyelectrolytes and small molecules electrolytes, which are combined with polar groups on the side chains are developed as effective interfacial materials [3,17]. They can be processed from alcoholic/water solvents, which make it possible to be deposited on top of the hydrophobic active layer to avoid intermixing with active layer corroding underlying layers. More importantly, the pendent compensating ions could form a strong interfacial dipole at the metal electrodes to tune the energy level alignment at the electrode/active layer interface, facilitating the charge extraction and collection. However, the mainstream

investigation on electrolytes for PSCs is still concerned on conjugated polyelectrolytes. Nowadays, some researches on small molecules conjugated electrolytes (SMCEs) also began to emerge and also drawn more and more attention for demonstrating excellent interfacial modification in the organic solar cells, owing to its high purity and easy adjusting structure [16,18–20]. Nevertheless, these categories of small molecule conjugated electrolytes which are explored as interfacial materials in PSCs remain limited and the relationship between structures of small molecule electrolytes and device performance is still unclear.

Herein, we developed three alcohol-soluble organic small molecule conjugated electrolytes as interfacial layers for I-PSCs, with the aim to uncovering the relationship between conjugated skeleton of SMCEs and photovoltaic performance. Three new SMCEs named 6,6',6'',6'''-(2,2'-bithiophene)-5,5'-diylbis(9H-diylbis(9H-fluorene-9,9,2-triyl))tetrakis(N,N,N-trimethylhexan-1-aminium) bromide (**2T-(FN-Br)**₂), 6,6',6'',6'''-(thiophene-2,5-diylbis(2-(5-octylthiophen-2-yl)-9H-fluorene-9,9,7-triyl))tetrakis(N,N,N-trimethylhexan-1-aminium) bromide (**3T-(FN-Br)**₂) and 6,6',6'',6'''-(2,2'-bithiophene)-5,5'-diylbis(2-(5-octylthiophen-2-yl)-9H-fluorene-9,9,7-triyl))tetrakis(N,N,N-trimethylhexan-1-aminium) bromide (**4T-(FN-Br)**₂), which have the same ammonium salt polar groups and only differ with conjugation length by varying the numbers of the thiophene bridges and thiophene terminal group, were presented Fig. 1a. Unlike the polar group, the conjugation length of the molecular skeleton could not cause a big fluctuation on the work function for electron extraction and collection. But enlarging the conjugation length, more uniform morphology, more intimate interfacial contact, and higher electron mobility can be easily achieved. Subsequently, by simply incorporation of thiophene units to enlarge π -delocalized conjugated skeletons, the device power conversion efficiency (PCE) could be readily boosted from 6.90% to 8.40%, based on polymer thieno[3,4-b]thiophene/benzodithiophene (PTB7) with [6,6]-phenyl C₇₁-butyric acid

methyl ester (PC₇₁BM) as active layer system (Fig. 1b).

2. Experimental section

2.1. Materials

All of the solvent and reagents were purchased from Alfa, Aldrich or other companies and used as received. 2-(7-Bromo-9,9-bis(6-bromohexyl)-9H-fluoren-2-yl)-5-octylthiophene, 5,5'-bis(9,9-bis(6-bromohexyl)-9H-fluoren-2-yl)-2,2'-bithiophene, 2,5-bis(9,9-bis(6-bromohexyl)-7-(5-octylthiophen-2-yl)-9H-fluoren-2-yl)thiophene and 5,5'-bis(9,9-bis(6-bromohexyl)-7-(5-octylthiophen-2-yl)-9H-fluoren-2-yl)-2,2'-bithiophene were synthesized as reported in the literatures [21,22].

2.2. Synthesis of the SMCEs

The synthesis routes toward the target SMCEs were outlined in Scheme S1. All the SMCEs shared the similar synthesis method. A large excess of trimethylamine (5.0 ml) 5,5'-bis(9,9-bis(6-bromohexyl)-9H-fluoren-2-yl)-2,2'-bithiophene (0.20 g, 0.175 mmol), 2,5-bis(9,9-bis(6-bromohexyl)-7-(5-octylthiophen-2-yl)-9H-fluoren-2-yl)thiophene (0.20 g, 0.174 mmol) and 5,5'-bis(9,9-bis(6-bromohexyl)-7-(5-octylthiophen-2-yl)-9H-fluoren-2-yl)-2,2'-bithiophene (0.20 g, 0.0130 mmol) in tetrahydrofuran (25 ml) at 0 °C with ice bath, respectively, then the mixture was allowed to warm up to room temperature gradually. The precipitate was redissolved by addition of excess water and an extra 2 ml trimethylamine was added at 0 °C. The resulting mixture was stirred for 24 h at room temperature and the crude yellow precipitate was collected by filtration. After purified by recrystallization from dichloromethane and hexane, a yellow solid was obtained. The synthesis details and

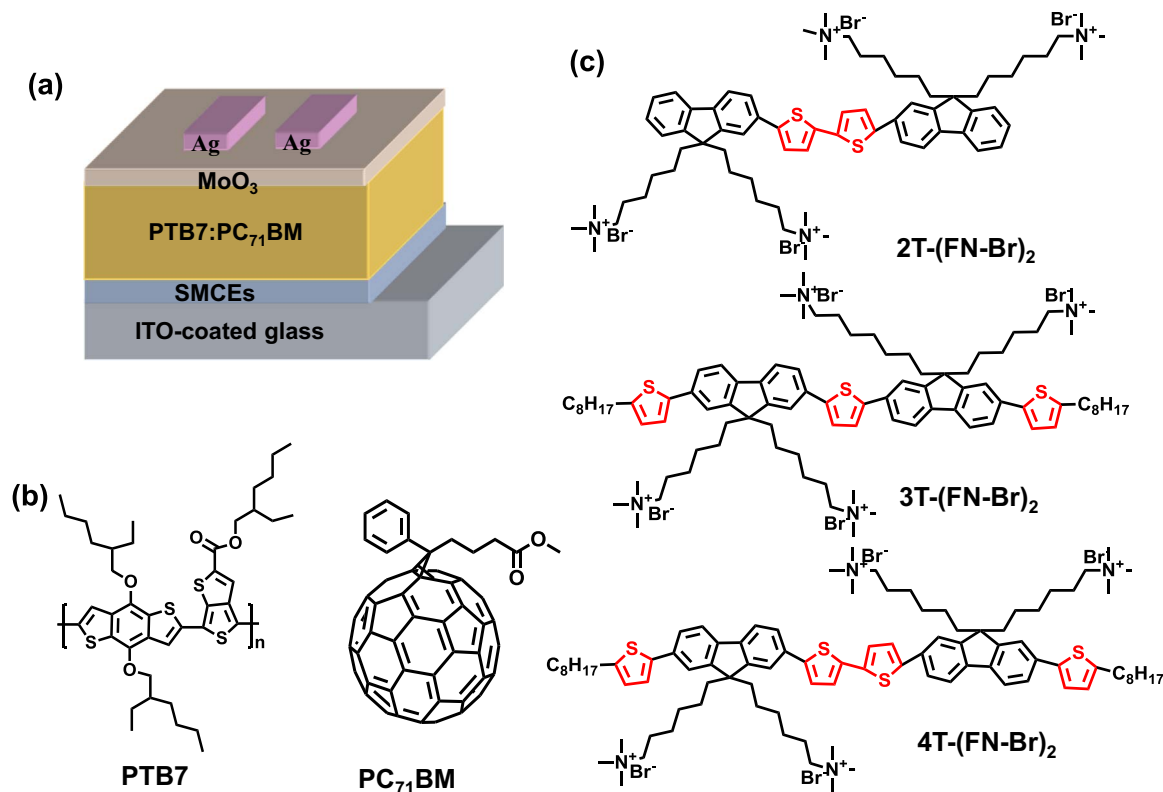


Fig. 1. (a) The configuration of I-PSC in the study, (b) Molecule structures of photoactive materials and (c) Molecule structures of 2T-(FN-Br)₂, 3T-(FN-Br)₂, 4T-(FN-Br)₂.

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