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Engineering the interconnecting layer for efficient inverted tandem polymer solar cells with absorption complementary fullerene and nonfullerene acceptors

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ABSTRACT

Applying materials with complementary optical absorption spectra to construct tandem solar cell can fulfill further improvement of photovoltaic performance, since tandem solar cell can harvest more photons and enhance the open-circuit voltage of the devices. For tandem solar cells, an interconnecting layer (ICL), which physically and electrically connects the front and the rear subcells, is essential to minimize the energy losses. Herein, we adopt MoO_3/Ag/PFN-Br as interconnecting layer to join front subcell of PBDTBDD:$PC_{70}BM$ and rear subcell of PTB7-Th:IEICO-4F for construction the inverted tandem solar cell with structure of ITO/TIPD/PBDTBDD:$PC_{70}BM$/MoO_3/Ag/PFN-Br/PTB7-Th:IEICO-4F/MoO_3/Al. The tandem devices demonstrate strong and wide light response in the wavelength between 300 and 1000 nm. A promising power conversion efficiency (PCE) of 9.74% has been achieved with an open circuit voltage (V_{oc}) of 1.503 V. The PCE of tandem cell is 41.36% higher than that (6.89%) of front subcell and 27.82% higher than that (7.62%) of rear subcell.

1. Introduction

Polymer solar cells (PSCs) have attracted increasing attention for their great potential of lightweight, mechanical flexibility, large area and low-cost fabrications [1–4]. Benefited from rational molecular design, morphological control, interfacial engineering, and optical manipulation, currently, the power conversion efficiencies (PCEs) of PSCs have been boosted to over 13% [5–7]. However, this efficiency is still lower than their inorganic (crystal silicon, CdTe and CIGs) and organic-inorganic hybrid perovskite counterparts. Since PSCs are exciton governed devices, and organic semiconductors suffer from low dielectric constants and limited charge-transporting properties, which limits the thickness of photoactive layer to be only around 100 nm to reduce carrier recombination [8]. Due to the significant carrier thermalization losses and incompletely absorption of the incident light, the maximum external quantum efficiency (EQE) of the PSCs is restricted to be ~ 70%, despite the internal quantum efficiency (IQE) of the photoactive layer can approach 100% [9], resulting in low short-circuit current density (J_{sc}) of the devices. In contrast, the single-crystal silicon is endowed with a broad optical absorption extended to 1100 nm [10,11], and the organic-inorganic hybrid perovskite bares high absorption coefficient within the whole visible range, rendering both devices demonstrate high J_{sc} [12,13]. Therefore, to further promote the performance of the PSCs, expanding the absorption spectra and enhancing the absorption coefficient of photoactive materials should be the effective methods. Fortunately, applying materials with complementary optical absorption spectra to construct tandem solar cell can fulfill further improvement of photovoltaic performance, since parallel connected tandem solar cell could enhance the J_{sc} by adopting complementary absorbers to harvest more photons and the open-circuit voltage (V_{oc}) could be enhanced by connecting two or more subcells in series [14,15]. In a tandem solar cell, the front subcell employs wide bandgap semiconductor material to capture photons in short wavelength and releases out a large V_{oc}, while the rear subcell adopts small bandgap semiconductor material to absorb photons in long wavelength and gives a relatively low V_{oc}, thus the carrier thermalization loss can be also reduced [16–18].

For tandem solar cells, an interconnecting layer (ICL), which physically and electrically connects the front and the rear subcells, is essential to minimize the energy losses [19–21]. Commonly, an ICL

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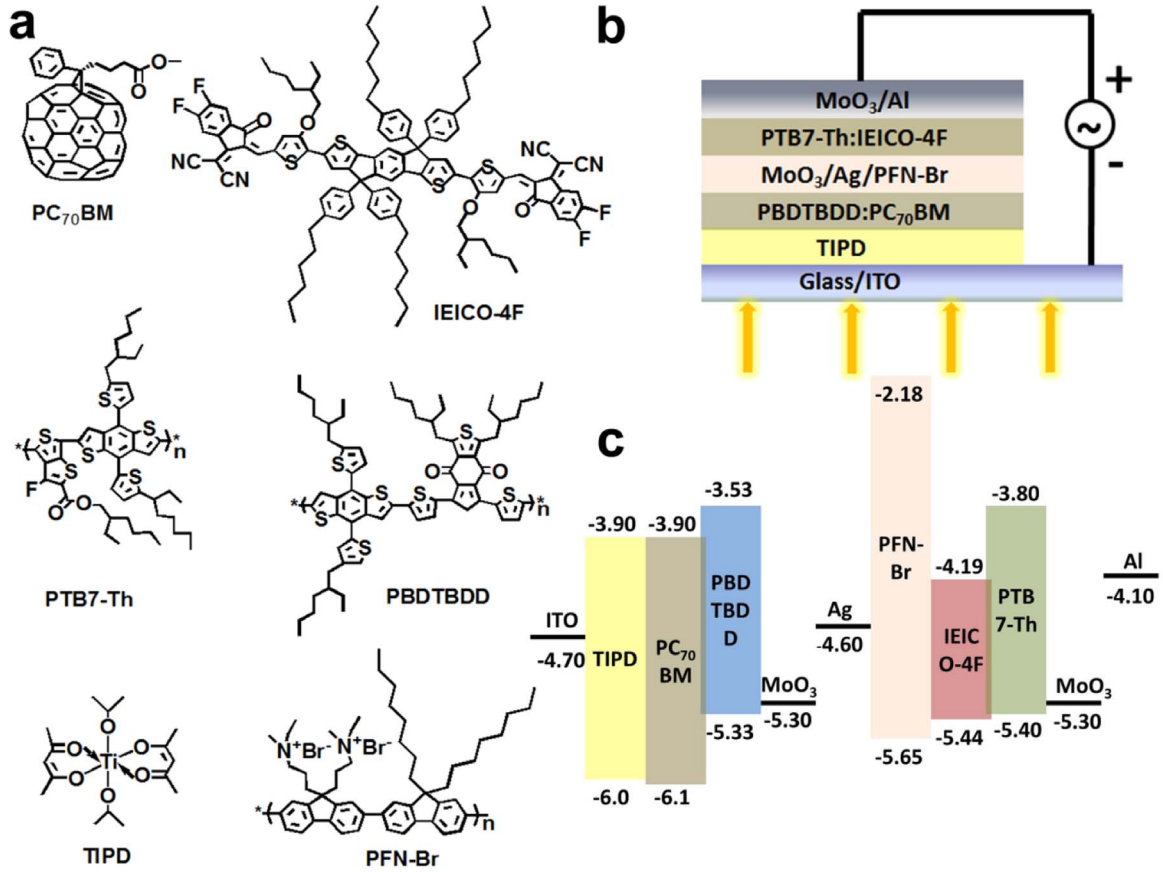

Fig. 1. (a) The molecular structures of $PC_{70}BM$, IEICO-4F, PTB7-Th, PBDTBDD, TIPD and PFN-Br; **(b)** Device structure of the inverted tandem solar cell; (c) Energy level diagram of inverted tandem solar cells.

consists of electron transport layer (ETL) to extract electrons from one subcell and hole transport layer (HTL) to extract holes from the other one. The ICL physically connected two subcells should be anti-solvent to protect the bottom subcell against the followed-up solution of the top subcell. Taken the inverted tandem solar cells as an example, though the exploration of ICLs has been continuously carried out, the widely reported ICLs in inverted tandem solar cells are typically with structure of HTL/ETL, typically poly(3,4-ethylenedioxythiophene) doped with polystyrene sulfonate (PEDOT:PSS)/ZnO [17], PEDOT:PSS/AgNPs/PEI [22], and PEDOT:PSS/PEIE [23,24]. Although these ICLs are endued with great optical and electrical properties, thermal annealing treatment (e.g. 120–150 °C) is required during preparation, which might influence the morphology of photoactive layer and potentially decrease the device performance. Occasionally, ICLs with structure of HTL/ultrathin metal (UTM)/ETL are also employed [25]. For example, introducing UTM (e.g. 0.5–10 nm Ag) with decent reflectivity could form micro cavity with another reflective electrode for fabricating highly efficient PSCs [26]. Since the micro cavities reinforce the optically confined incident lights, comparable or even higher PCE can be achieved in compared with those ITO-based devices [27]. In tandem solar cells, the UTM is commonly functioned as carrier recombination sites to increase the holes and electrons recombination chance. Furthermore, the resonant light in an optical micro cavity can be manipulated by varying the layer thickness within the tandem device, especially the thickness of the photoactive layer. MoO_3/Ag/PFN (poly[(9,9-bis(3′-((*N*,*N*-dimethyl)-*N*-ethylammonium)-propyl)-2,7-fluorene)-*alt*-2,7-(9,9-dioctylfluorene)]) has been used as ICL in tandem solar cells, by introducing ultrathin Ag layer, the J_{sc} of the tandem solar cell is greatly enhanced due to the formation of micro cavity to capture more light [28]. The previous reports demonstrate that PFN-Br works very well as ETL in nonfullerene acceptor based PSCs [29,30]. However, there is no report about applying PFN-Br as the ETL in inverted PSCs. The possible reason should be that the energy level of PFN-Br does not match with that of the ITO electrode. As for UTM, another important function is tuning the work function. Therefore, combined with the UTM, PFN-Br should exhibit acceptable property in inverted PSCs and could further be applied as ICL in inverted tandem solar cells.

Herein, we utilize MoO_3/Ag/PFN-Br as ICL, 5,7-bis(2-ethylhexyl) benzo[1,2-c:4,5-c′]dithiophene-4,8-dione (PBDTBDD):phenyl-C_{70}-butyric acid methyl ester ($PC_{70}BM$) for front subcell and poly[4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b:4,5-b']dithiophene-*co*-3-fluorothieno[3,4-b]thiophene-2-carboxylate] (PTB7-Th):2,2′-((2*Z*,2′*Z*)-(((4,4,9,9-tetrakis(4-hexylphenyl)-4,9-dihydro-sindaceno[1,2-b:5,6-b']dithiophene-2,7-diyl)bis(4-((2-ethylhexyl)oxy)thiophene-5,2-diyl))bis(methanylylidene))bis(5,6-difluoro-3-oxo-2,3-dihydro-1H-indene-2,1-diylidene))dimalononitrile (IEICO-4F) for rear subcell to construct inverted tandem solar cells with complementary absorption spectra from 300 to 1000 nm. IEICO-4F is a recently developed nonfullerene acceptor with absorption spectra extending to 1000 nm [31], which should be a promising alternative material in tandem solar cells. In the rear subcell, the Ag/PFN-Br is successfully used as the cathode buffer layer where Ag plays a role in aligning the energy level of ITO and PFN-Br. By carefully optimizing the both photoactive layer thickness of front and rear subcells as well as tuning the ICL, a desirable PCE of 9.74% and a promising V_{oc} of 1.503 V have been achieved for tandem devices.

2. Experimental section

2.1. Materials

Etched indium tin oxide glass (ITO) glass (15 Ω/□) was purchased from CSG PV Tech Co., Ltd (China). Titanium (diisopropoxide) bis (2, 4-