



Enhancing the performance of planar heterojunction perovskite solar cells using stable semiquinone and amine radical modified hole transport layer

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HIGHLIGHTS

- Dopamine radical enhanced the PCE and device durability.
- PEDOT with high work function was obtained via semiquinone containing backbone.
- The HTL with excellent hole transport and efficient electron blocking.
- The PCE over 15.3% was achieved for PSCs with a simple planar heterojunction structure.

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ABSTRACT

Highly efficient organic-inorganic hybrid planar heterojunction perovskite solar cells with an architecture of indium tin oxide (ITO)/PEDOT:PSS-NH₂-OH/CH₃NH₃PbI₃/PCBM/Al are fabricated *via* one-step, solution process by employing the dopamine modified PEDOT:PSS (PEDOT:PSS-NH₂-OH) as the hole transport layer, of which the power conversion efficiency of 15.34% is achieved with negligible hysteresis regardless of different scanning directions and scanning speeds. The average efficiency of twenty devices with PEDOT:PSS-NH₂-OH is increased to 14.16% from 10.67% with PEDOT:PSS, and the former devices exhibit good reproducibility with small standard deviations. Cyclic voltammetry results show that the highest occupied molecular orbital energy level of PEDOT:PSS-NH₂-OH (5.32 eV) matches well with the valence band of CH₃NH₃PbI₃ absorbers (5.4 eV). Photoluminescence measurements indicate that the CH₃NH₃PbI₃ film deposited on PEDOT:PSS-NH₂-OH has strong charge extraction capability than that on PEDOT:PSS. Electrochemical impedance spectroscopy results suggest that the photovoltaic devices exhibit efficient hole transport and excellent electron blocking employing PEDOT:PSS-NH₂-OH as the hole transport layer, as well as have low series, contact resistance and large recombination resistance. The research demonstrates that the stable semiquinone and amine radical modified PEDOT:PSS can acts as an outstanding hole transport material for fabricating high-efficient perovskite solar cells.

1. Introduction

Organic-inorganic hybrid perovskite solar cells (PSCs) as a hotspot in academia in recent years have captured enormous attention owing to the great superiorities of perovskite materials, such as ambipolar properties, direct band gap, strong absorption, high carrier mobility,

long carrier diffusion, low exciton binding energy, *etc.* [1–4]. There has been an awe-inspiring breakthrough in photovoltaic community by multidisciplinary efforts, resulting in the state-of-the-art of power conversion efficiency (PCE) of PSC up to 22.7% [5]. Mesoscopic structural and planar-based PSCs have been widely studied in past years. Planar heterojunction (PHJ) PSCs, especially inverted PHJ-PSCs,

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have been attracting much attention due to the simple architecture and inconspicuous hysteresis, as well as the low-cost, low-temperature, solution processing and matching with high-output, large-scale roll-to-roll printing techniques [6–9].

It is well recognized that the active layer of photovoltaic devices absorb sunlight and produce electrons and holes separated, which then transport to electron transport layer (ETL) and hole transport layer (HTL), respectively, and ultimately are collected by the electrodes. Efficient carrier extraction and transport play the key role on device performance parameters such as open-circuit voltage (V_{oc}), PCE and hysteresis [10–14]. The poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonic acid) (PEDOT:PSS) is one of the main HTL materials employed in solar cells [15,16]. But, due to its intrinsic properties of acidity and hygroscopicity, it is unfavorable to achieve long-term stability. As a good HTL, it not only needs high electrical conductivity and excellent optical transparency with wide bandgap, but also it should be an outstanding electron blocking property with preeminent hole transport. Many studies have been done for probing alternative HTL materials, for instance, metal oxides MoO_3 [15,16], WO_3 [17,18], V_2O_5 [19], CuI [20], NiO_x [21–23], PbS quantum dots [24], $CuSCN$ films [25] and so on.

Motivated by our previous work on the stable semiquinone radical and efficient modification of ITO work function [10], we systematically investigated the effects of dopamine (DA) doping on electron donating capability of PEDOT:PSS and perovskite film quality as well as the charge recombination kinetics in the solar cells. In comparison with the pristine PEDOT:PSS, we found that DA doping endows PEDOT:PSS with higher radical content and stronger charge extraction capability from perovskite to HTLs [11]. PEDOT:PSS-NH₂-OH with typical semiconductor behavior exhibits more efficient electron donating effect than PEDOT:PSS due to the further doping of DA-PEDOT:PSS on perovskite layer during device fabrication, thereby providing better trap passivation capability. Besides improving perovskite film crystallinity, DA with amino and hydroxyl groups interacts with the under-coordinated Pb atoms on the perovskite crystal, resulting in the reduced surface recombination. Due to the electron transfer process of phenol-quinone-PEDOT backbone, PEDOT:PSS-NH₂-OH with deep work function (-5.32 eV) exhibits better capability to suppress the interfacial trap-assisted recombination with longer charge recombination lifetime and higher charge extraction rate. Eventually, the V_{oc} , FF and PCE of the PEDOT:PSS-based devices simultaneously increased from 0.86 V, 69.3% and 12.01% to 0.97 V, 78.15% and 15.34% when PEDOT:PSS-NH₂-OH was introduced.

Our molecular design was presented in Scheme 1. The amino and hydroxyl radical dual-functionalization of DA endows PEDOT:PSS with several excellent properties: 1) High radical content and strong charge extraction capability from perovskite to HTL. 2) Desirable energy level alignment with perovskite. 3) Favorable interaction with perovskite for high-quality film formation. 4) Efficient surface trap passivation effects for perovskite film with a long charge recombination lifetime and high charge extraction rate. With these multiple functions, it was succeed to apply in perovskite $CH_3NH_3PbI_3$ (MAPbI₃) photovoltaic devices with a structure of ITO/PEDOT:PSS-NH₂-OH/MAPbI₃/PCBM/Al that fabricated by one-step, solution-processing under low temperature. As a result, encouraging results with sharply enhanced performance (a PCE of 15.34%) were achieved as compared to PEDOT:PSS-based PHJ-PSCs (a PCE of 12.01%) with negligible hysteresis regardless of scanning directions and scanning speeds. These in-depth understanding of structure-performance clearly provide new guideline to design effective HTLs inspired by the electron transfer of semiquinone radical, phenol derivatives for further improvements of inverted PHJ-PSCs.

2. Experiment section

2.1. Materials

Methylammonium iodide (CH_3NH_3I , MAI, 99%) and lead iodide (PbI_2 , 99%) were purchased from Wuhan Jingge and Shanghai Zhengpin, respectively. N,N -Dimethylformamide (Super dry, DMF) and chlorobenzene (Super dry, CB) were provided by J&K Seal. Fullerene derivative [6,6]-phenyl-C61-butyric acid methyl ester (PCBM, Dye Source, American) was supplied by American Dye Source. PEDOT:PSS (PVP AI 4083) was purchased from Baytron.

2.2. Preparation of PEDOT:PSS-NH₂-OH

PEDOT:PSS-NH₂-OH was synthesized as following detailed process: 20 g polystyrene sulfonic acid (PSS, Mw = 75000 Da, 30 wt.%) and 1 g of 3,4-ethylenedioxy thiophene (EDOT) and was mixed, and the solution was adjusted to pH = 2. Then, PEDOT:PSS-NH₂-OH (1% in water, v/v) was prepared by adding the product above into the PEDOT:PSS-4083 solution with the ratio of 1:5 (v/v). Dopamine-copolymerized PEDOT:PSS was obtained and studied carefully. The molecular structure, UV-vis absorption properties of modified PEDOT:PSS were also investigated. The molecular structure of PEDOT:PSS-NH₂-OH is proposed and shown in Scheme 1. All the chemicals were used as received without further purification.

2.3. Precursor preparation

The perovskite precursor was prepared by dissolving MAI and PbI_2 at a molar ratio of 1:1 in anhydrous DMF at the concentration of 550 mg/ml. PCBM was dissolved in anhydrous CB with a concentration of 15 mg/ml. Perovskite precursor solution and PCBM solution were energetically stirred over night at 60 °C to ensure dissolve adequately. A 0.22 μ m PVDF filter was employed to filter the perovskite precursor before deposition.

2.4. Device fabrication

The inverted PHJ-PSC with the structure of ITO/HTL/MAPbI₃/PCBM/Al were fabricated in this study. The patterned indium tin oxide (ITO) glass substrate was ultrasonically cleaned in acetone, detergents, distilled water and isopropyl alcohol for 20 min, respectively. After the cleaning and drying by nitrogen flow, the ITO-coated substrate was treated using UV-ozone for 20 min to reform and removal of organic substance. The HTL materials PEDOT:PSS and PEDOT:PSS-NH₂-OH were spin-coated on the ITO substrate at a spinning speed of 3000 rpm for 30 s and 4000 rpm for 30 s, and then annealed on hot plate at 150 °C for 15 min and 110 °C for 10 min, respectively, resulting in a thickness of about 35 nm for PEDOT:PSS and 30 nm for PEDOT:PSS-NH₂-OH. It is noted that the PEDOT:PSS solution was agitated for 20 min and PEDOT:PSS-NH₂-OH solution was ultrasonically for 10 min before spin-coating. The solvent-induced-fast-crystallization deposition method [14,27–29] was used to fabricate the perovskite layer to obtain dense and smooth high-quality perovskite film in a nitrogen-filled glovebox (both H₂O and O₂ < 1.0 ppm), and ca. 300 nm thick of red brown MAPbI₃ perovskite film can be obtained ultimately. PCBM solution was spin-coated onto perovskite layer at a speed of 3000 rpm for 30 s, resulting in a thickness about 30 nm. Finally, a 100 nm Al electrode was deposited by thermal evaporation under a vacuum of about 8.0×10^{-6} mbar, the active area of the devices is 0.09 cm² that determined by the size of Al pads.

2.5. Characterization

The absorption spectra, crystallographic properties, morphologies of perovskite films were characterized by employing ultraviolet-visible

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