



Performance enhancement of perovskite solar cells through interfacial engineering: Water-soluble fullereneol C₆₀(OH)₁₆ as interfacial modification layer

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ABSTRACT

TiO₂ is widely used in conventional *n-i-p* type perovskite solar cells (Pero-SCs) as electron transporting layer (ETL). However, its drawbacks such as photocatalytic properties, low electron mobility, and a process of high temperature sintering, hindered the further improvement of device performance. In this work, a vacuum-deposited fullerene C₆₀ was utilized as ETL, and a easily accessible fullerene derivative, C₆₀(OH)₁₆ fullereneol, was employed between ITO electrode and C₆₀ ETL to modify ITO for the first time. After introducing the C₆₀(OH)₁₆ fullereneol layer, the Pero-SC obtained the favorable energy level alignment between ITO cathode and perovskite layer, and the device based on C₆₀(OH)₁₆/C₆₀ ETL achieved a champion PCE of 16.70%, which showed a 15.41% enhancement compared to the Pero-SC with pristine C₆₀ ETL (14.47%). The mechanisms of performance improvement were systemically discussed with respect to the optimized energy level, efficient charge transport and reduced interfacial resistance. This work exhibited that fullereneol C₆₀(OH)₁₆ is a promising interfacial modification material for the *n-i-p* type Pero-SCs.

1. Introduction

In recent years, organic-inorganic metal halide perovskite solar cells (Pero-SCs), which is one of the most promising candidates in the field of photovoltaic technology, have attracted extensive attentions worldwide [1,2]. Since it was first reported in 2009, in only a few years the Pero-SCs developed rapidly with its power conversion efficiency (PCE) skyrocketed from 3.8% to 22.7% [3–6]. This unprecedentedly rapid development of Pero-SCs is ascribed to its low manufacturing costs, high efficiency of solar conversion and excellent photoelectric properties such as long exciton diffusion length and exciton lifetime, high absorption coefficient and better carrier mobility [7–11].

The rapid progress of PCE for the Pero-SCs is mainly benefited from the improvement of high quality perovskite film with large grain size and smooth surface morphology, as well as the development of efficient electron transporting materials (ETM) and hole transporting materials (HTM) [12–16]. The planar Pero-SCs have two major types of *n-i-p* and *p-i-n* structures. For the conventional *n-i-p* type Pero-SCs, the most commonly used ETM is compact TiO₂ (c-TiO₂) [17]. However, some

drawbacks of TiO₂ have limited the application of TiO₂ as electron transport layer (ETL) in Pero-SCs [18,19]. For example, the low electron mobility of TiO₂ could lead to charge accumulation in the layer. TiO₂ also needs an additional step of sintering at 500 °C to form a crystalline phase. Further, its photocatalytic properties could result in light instability of the Pero-SCs when exposed TiO₂ in UV light. Therefore, to further obtain stable power output, suppress hysteresis, facilitate the electron injection and improve PCE of the Pero-SCs, tremendous efforts were devoted to the development of novel ETMs (such as WO₃, SnO₂, ZnO and ionic liquid) to replace or modify TiO₂ [19–23].

Fullerenes and their derivatives have great potential of application as the ETL or modification of the Pero-SCs with the advantage of efficient electron transporting, the ability to reduce hysteresis and the simple film formation method [24–32]. Fullerene derivative could be employed to modify the TiO₂ layer, which could passivate the deep trap states of TiO₂ and enhance the efficiency and stability of Pero-SCs [33]. These results demonstrate the great potential of fullerene and their derivatives for the application as ETM to improve the performance of Pero-SCs. Fullereneol, an easy-accessible polyhydroxylated fullerene

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derivatives, has excellent electron mobility and good water solubility. In recent reports, fullerene has been proven to be an efficient interfacial material in solar cells, Yang et al. applied fullerene as a cathode buffer layer in inverted organic solar cells, with which the PCE of the inverted device was enhanced [34]. In our recent study, a fullerene $C_{60}(OH)_{24}$ was used to modify TiO_2 ETL in *n-i-p* type Pero-SCs, after the modification of $C_{60}(OH)_{24}$, the device demonstrated a facilitated charge extraction and the device performance showed an enhancement of 17.5% compared to the control device with pristine TiO_2 ETL [35]. Thus, these encouraging results propelled us to explore the application of fullerene in Pero-SCs with variable structures.

Herein, we reported the application of a fullerene $C_{60}(OH)_{16}$, synthesized by a simple and solvent-free method at the room temperature, as interlayer between ITO and C_{60} in *n-i-p* Pero-SCs. The devices without and with $C_{60}(OH)_{16}$ fullerene layer were fabricated and the systematic characterizations were carried out to investigate the effect of $C_{60}(OH)_{16}$ fullerene layer on photovoltaic performance of the Pero-SCs. The results show that, after introducing $C_{60}(OH)_{16}$ fullerene layer modified ITO under C_{60} ETL, the device obtained the optimized interface energy level alignment and reduced the energy loss in electron transporting process. Furthermore, the Pero-SCs demonstrated a facilitated electron transfer efficiency and a decreased transport resistance with the modification of $C_{60}(OH)_{16}$ fullerene layer. While the control device without $C_{60}(OH)_{16}$ fullerene only reached a PCE of 14.47%, the Pero-SCs with $C_{60}(OH)_{16}$ fullerene layer presented a notable enhanced PCE of 16.70%.

2. Experimental section

2.1. Materials

Fullerene C_{60} (99.9%) was purchased from Dade Carbon Nanotechnology Co., Ltd, H_2O_2 (30 wt%) was bought from Aladdin Industrial Corporation, NaOH (AR) and methanol were purchased from Sinopharm Chemical Reagent Co., Ltd and Chinasun Specialty Products Co., Ltd, respectively. And all these samples are used without further purification.

We synthesized Methylammonium iodide (MAI) in our laboratory according to the literature [36]. PbI_2 (99.999%) was bought from Alfa Aesar, Methylammonium Chloride (MACl) was purchased from Xi'an Polymer Light Technology Crop. P3HT was obtained from Rieke Metals, Inc.

2.2. Device fabrication

ITO substrates were sequentially cleaned 20 min by ultrasonic in detergent, deionized water, acetone, ethanol and isopropyl alcohol. After being dried at 150 °C for 10 min, the ITO glass was treated for 20 min with ultraviolet ozone (UVO) to increase affinity for water molecules. The $C_{60}(OH)_{16}$ was dissolved in deionized water at four different concentrations of 0.3 mg/mL, 0.5 mg/mL, 0.7 mg/mL and 1 mg/mL. And then these solutions were spin-coated on ITO substrates at 1500 rpm for 50s. The substrates were transferred into the glove box with nitrogen atmosphere after annealed at 120 °C for 20 min. 12 nm of C_{60} was deposited on top of the $C_{60}(OH)_{16}$ fullerene using the method of vacuum evaporation.

461 mg PbI_2 was dissolved in 1 mL N, N-dimethylformamide (DMF, 99.8%, J&K), 50 mg MAI and 5 mg MACl (mass ratio of 10:1) were dissolved in 1 mL isopropanol (IPA, J&K). All the precursor solutions were placed on a hot plate at 70 °C and stirred for 12 h. These precursor solutions were filtered by 0.45 μ m polytetrafluoroethylene (PTFE) filter before use. The perovskite active layer was deposited using the method of sequential two-step deposition. 40 μ L of PbI_2 solution was spin-coated on the top of C_{60} at 4500 rpm for 45 s. During this process, additional 40 μ L of the mixed solution of MAI: MACl (10:1) was further dropped on the spinning substrate 25 s prior to the end of this PbI_2

coating procedure. Thermal annealing for the film was processed at 100 °C for 3 min in a nitrogen atmosphere. Perovskite films turned dark immediately after annealing.

30 mg P3HT as a hole transport material was dissolved in 1 mL *o*-dichlorobenzene, which added with 20.4 μ L bis(trifluoromethane) sulfonamide lithium salt (Li-TFSI) solution (28.3 mg Li-TFSI in 1 mL acetonitrile) and 10.2 μ L 4-tertbutylpyridine (t-BP). P3HT HTL was spin-coated on the top of perovskite at 1500 rpm for 60 s. 7 nm of MoO_3 was deposited by vacuum evaporation as buffer layer. Finally, 80 nm of Ag was vacuum evaporated as a counter electrode using a mask plate. 0.0757 cm^2 of measuring active area was defined by a shadow mask. The configuration of ITO/ $C_{60}(OH)_{16}$ / C_{60} /Au was used to measure the direct current conductivity. 80 nm Au was vacuum evaporated and using a shadow mask with an active area of 0.064 cm^2 .

2.3. Materials and devices characterization

The instrument of Bruker VERTEX 70 V (Germany) was used to obtain Fourier transform infrared spectrometer (FTIR) spectra. Thermal gravimetric analysis (TGA) was obtained with a PerkinElmer Pyris 6. Work function (WF) was measured by Ultraviolet photoelectron spectroscopy (UPS, Escalab 250Xi, Thermo Fisher, USA). Current density–voltage ($J-V$) curves of a perovskite solar cells (Pero-SCs) were acquired by applying an external voltage bias and using Keithley 2400 digital source meter to record the current response. A Xenon arc lamp with power of 100 mW/cm^2 simulated Air Mass 1.5 global (AM 1.5G) solar illumination. Steady-state photocurrents under applied voltage were obtained in the same instrument. The measurements of $J-V$ and steady-state photocurrents were carried out in glove box. Incident photo-to-current conversion efficiency (IPCE) of Pero-SCs was measured using Enli Technology Co., Ltd. QE-R3011 in air without bias light. Standard commercial single-crystal silicon solar cells were used to corrected illumination intensities. Using spectroscopic ellipsometer (M-2000 V, J. A. Woollam Co., USA) measure the thicknesses of fullerene films. SU8010 (Hitachi produced) was used to acquire scanning electron microscopy (SEM) images. X-ray diffraction (XRD) patterns (2 θ scans) were recorded by X-ray diffractometer (D2 PHASER, Bruker, Germany) from 10° to 70° with radiation source angle $\lambda = 1.54184 \text{ \AA}$. Ultraviolet-visible (UV-vis) spectra were carried out using an UV-vis spectrophotometer (Cary 6000, Agilent). Steady-state photo-luminescence (PL) were measured on FLS980 (Edinburgh Instrument, UK) with excitation wavelength is 470 nm. Time-resolved PL spectra were acquired on Lifespec (Edinburgh Instrument, UK) with a 477 nm laser (5 MHz). The alternating current impedance spectrometry (ACIS) was obtained in dark condition using IM6 electrochemical workstation (Zahner Zennium, Germany). The bias voltage is 1.0 V and the frequency ranged from 0.1 Hz to 4 MHz. The effective area of the cell is 0.1842 cm^2 . The impedance spectra was fitted by Z-view software to obtain the impedance parameters.

3. Results and discussion

Water-soluble fullerene $C_{60}(OH)_{16}$ was conveniently synthesized by a solvent-free reaction of fullerene [37]. The details of synthetic process were described in Supporting Information (SI). As presented in Fig. 1a, $C_{60}(OH)_{16}$ fullerene was prepared by C_{60} fullerene with a mixture of H_2O_2 and NaOH under grinding conditions at room temperature in the air. The FTIR spectrum of $C_{60}(OH)_{16}$ was presented in Figure S1 in SI, in which a strong peak centered at 3409 cm^{-1} was assigned to ν O-H, and the characteristic vibration bands of ν C-O, δ_s C-O-H, and ν C=C located at 1090 cm^{-1} , 1376 cm^{-1} and 1610 cm^{-1} , respectively [35,37]. These results are similar to the FTIR spectra of other fullerene reported before and confirmed that -OH groups have been successfully attached to the C_{60} cage [38–40]. The FT-IR of fullerene film before and after being heated to 120 °C was measured to further investigate the thermal stability of fullerene for annealing treatment. Figure S2 presented similar

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