

Improving the efficiency of $\text{CH}_3\text{NH}_3\text{PbI}_3$ based photovoltaics by tuning the work function of the PEDOT:PSS hole transport layer

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Abstract

The open-circuit voltage of $\text{CH}_3\text{NH}_3\text{PbI}_3$ based photovoltaics was enhanced from 0.77 V to 0.9 V, which resulted in an improvement of the average power conversion efficiency from 5.63% to 11.74% achieved by tuning the work function of the poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) hole transport layer. The higher work function of the PEDOT:PSS (1:20 wt%) thin film, as compared to the PEDOT:PSS (1:6 wt%) and PEDOT:PSS (1:2.5 wt%) thin films, was observed using photoelectron spectrometer. The carrier density of PEDOT was obtained by analyzing the transmittance spectrum of PEDOT:PSS/glass with the transfer matrix method. The Drude model can be used to explain the increased work function of the PEDOT:PSS thin film obtained by decreasing the ratio of PEDOT to PSS. The increased carrier density of the PEDOT in PEDOT:PSS thin films originated from conformational changes in the PEDOT chains as confirmed by Raman scattering spectroscopy.

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1. Introduction

In the past three years, organometal halide perovskite ($\text{CH}_3\text{NH}_3\text{PbI}_3$ or $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$) based photovoltaics (PVs) have been intensively investigated. A high power

conversion efficiency (PCE) can be achieved using solution processes under low temperatures ($<120^\circ\text{C}$). To date, the highest reported PCE for a $\text{CH}_3\text{NH}_3\text{PbI}_3$ based PV is 19.3%, obtained by engineering the work functions of the electron transport layer (ETL) and hole transport layer (HTL) Zhou et al., 2014. There are several reasons which can be used to explain the high PCE of $\text{CH}_3\text{NH}_3\text{PbI}_3$ based PVs. Their relatively low refractive index ($n \sim 2.7$) and high extinction coefficient ($k \sim 0.3$) are advantageous for light

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harvesting (Lin et al., 2015a,b; Ziang et al., 2015) without significant reflection from the interface between the substrate and the $\text{CH}_3\text{NH}_3\text{PbI}_3$. The long wavelength absorption edge approximately located at about 780 nm (1.59 eV) Yamada et al., 2014; Aharon et al., 2014; Anaya et al., 2015 can absorb half of the incident sunlight. The small exciton binding energy of 50 meV (Tanaka et al., 2003; D'Innocenzo et al., 2014) means that the excitons can be efficiently dissociated at the interface between the perovskite and the ETL (HTL) Stranks et al., 2013. In addition, the high electron and hole mobilities originating from the low Urbach energy of 15 meV (Wolf et al., 2014; Lee et al., 2015) and the photoferroic effect (Forst et al., 2014) result in a high fill factor ($\text{FF} > 0.7$). The first perovskite based PV was fabricated using the following structure: FTO/Pt/electrolyte/perovskite/meso- TiO_2 /FTO/glass (Kojima et al., 2009). The PCE of perovskite based PVs could be improved from 3.8% (Kojima et al., 2009) to 17.1% (Im et al., 2014) by using an optimized thickness of the mesoporous TiO_2 (Heo et al., 2013). The main drawback is that the sintering temperature for anatase TiO_2 has to be higher than 450 °C (Schattauer et al., 2012; Zhang et al., 2015) which can damage the plastic substrate. For low temperature fabrication, a ZnO nanoparticle film can be used to replace the sintered TiO_2 thin film as the ETL. The optimized thickness of a ZnO nanoparticle film for a perovskite based PVs is about 25 nm, producing a high PCE of 15.7% (Liu and Kelly, 2014). With this flexible plastic substrate, the PCE of ZnO-based perovskite PVs decreases from 15.7% to 8.6% (Kumar et al., 2013). P-type poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) thin film can be used as the HTL for realizing flexible perovskite PVs while maintaining the PCE (You et al., 2014). The structure of the flexible PV is Al/PCBM/perovskite/PEDOT:PSS/ITO/PET, where phenyl- C_{61} -butyric acid methyl ester (PCBM) is used as the ETL, and polyethylene terephthalate (PET) is used as the substrate. A high PCE of 17.7% can be achieved by using the following structure: Al/PCBM/perovskite/PEDOT:PSS/FTO/glass (Nie et al., 2015) when the grain size of the perovskite is 180 μm . However, the PCE is limited by the open-circuit voltage (V_{OC}) which is equal to the difference in the Fermi level between Ag (−4.23 eV) and PEDOT:PSS (−5.1 eV) (Yip and Jen, 2012). The use of an Al/Ca cathode electrode and phenyl- C_{71} -butyric acid methyl ester (PC_{71}BM) can improve the V_{OC} of perovskite based PVs by 0.04 V (Chiang et al., 2014), but Ca is easily oxidized, which leads to degradation of the PCE. In reference Lim et al. (2014), the use of a larger work function HTL for perovskite based photovoltaics leads to a better alignment with the perovskite valence band. Therefore, the tuning of the Fermi level (work function) of a PEDOT:PSS thin film is another way (Lee and Chung, 2008) to improve the V_{OC} of perovskite based PVs. The PEDOT:PSS (1:6 wt%) thin film is widely used as the HTL in organic PVs (Yip and Jen, 2012) and perovskite based PVs (Jeng et al., 2013; Liang et al., 2014; Bi et al.,

2014a,b; Jung et al., 2014). The PEDOT:PSS (1:2.5 wt%) thin film with an immersion treatment can replace the ITO thin film for a standalone anode electrode (Chang et al., 2014a,b), but the V_{OC} of the organic PVs is reduced from 0.63 V to 0.54 V due to the smaller work function of the PEDOT:PSS (1:2.5 wt%) thin film. In this study, a PEDOT:PSS (1:20 wt%) thin film was used as the HTL to improve the V_{OC} of $\text{CH}_3\text{NH}_3\text{PbI}_3$ based PVs from 0.77 V to 0.90 V. Square wave voltammetry (SWV) and photoelectron spectrometer were used to determine the Fermi levels and work functions of PEDOT:PSS thin films, respectively. Transmittance and Raman scattering spectra were used to explore the origins of the tunable Fermi level (work function) in the PEDOT:PSS thin films.

2. Experiments

In our device fabrication process, the PEDOT:PSS thin films were spin-coated onto transparent conductive ITO glass (UR-ITO007-0.7 mm, from Uni-onward Corp. Taipei, Taiwan) with a sheet resistance of 10 Ω/sq then subjected to thermal annealing at 120 °C for 10 min, to act as the HTL. PEDOT:PSS (1:2.5 wt%) (PH1000), PEDOT:PSS (1:6 wt%) (AI4083), and PEDOT:PSS (1:20 wt%) (CH8000) were purchased from Heraeus Ltd. Then, PbI_2 (Aldrich) and $\text{CH}_3\text{NH}_3\text{I}$ were dissolved in a γ -butyrolactone (GBL)/dimethylsulfoxide (DMSO) mixture (1:1 v/v) to be used as the perovskite precursor. $\text{CH}_3\text{NH}_3\text{I}$ powder was synthesized using a method described in literature Etgar et al. (2012). The perovskite precursor was spin-coated on top of the PEDOT:PSS/ITO/glass. The spinning process was divided into two steps which included a slow spin rate of 1000 rpm for 10 s and a fast spin rate of 5000 rpm for 20 s. Toluene (50 μL) was dropped at the last 2 s during the spin-coating process to form the perovskite precursor (Jeon et al., 2014; Lin et al., 2015a,b). After that, the sample was thermally annealed at 100 °C for 5 min to produce a 260-nm thick perovskite film. Then, a 50-nm thick PCBM film was spin-coated on top of the perovskite film with solvent annealing under a *o*-dichlorobenzene vapor environment for 20 min. Finally, a 80-nm-thick Ag film was evaporated on top of the PCBM, to act as the cathode electrode.

The active area of the PV device was 0.1 cm^2 . The current density–voltage (J – V) curves were measured using a Keithley 2400 source-measurement system. The intensity of the simulated sunlight was calibrated using an NREL-certified Si solar cell (Oriel, 91150V) with a KG-5 colored glass bandpass filter to have an intensity of 100 mW/cm^2 (AM 1.5G). The surface morphologies and phase images of the PEDOT:PSS thin films were analyzed by an atomic force microscope (Seiko SPA-300HV). SWV was performed to estimate the Fermi levels of the PEDOT:PSS thin films. The resolution of SWV is 10 mV. SWV experiment was conducted using a three-electrode configuration with an ITO working electrode. A platinum wire was used as the auxiliary electrode and a home-made Ag/AgCl, KCl

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