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Efficient polymer photovoltaic cells using solution-processed MoO_3 as anode buffer layer

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ABSTRACT

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1. Introduction

Organic photovoltaic (PV) cells have been the hot research topic in recent years due to their advantages of low-cost manufacturing, light weight and good flexibility [1-5]. Among these PV cells, solution-processed bulk heterojunction PV cells with donor-acceptor (D-A) blends sandwiched between the anode and cathode are the most promising alternative to realize large-scale production. In bulk heterojunction PV cells, the charges generated in the active layer upon light absorption are transported via D-A networks and collected by the electrodes to realize light-electricity conversion. Usually, the indium-tin oxide (ITO) anode needs to be modified to enhance the device performance of the polymer PV cells. Note that in the simplest bulk heterojunction cell architecture with the active layer sandwiched between the anode and cathode, the active layer donor and acceptor materials are both in direct contact with the anode, and it is possible for the acceptor material to transfer electrons to the hole-collecting anode, thereby resulting in large leakage current and decreased PV performance of the polymer PV cells. To tackle these problems, interfacial buffer layers are always inserted between the active layer and electrodes to enhance the collection of the photogenerated charges and decrease the leakage current [6-8]. Poly(ethylenedioxythiophene):poly (styrenesulfonate) (PEDOT:PSS) is commonly adopted as the anode buffer layer in traditional polymer PV cells [6-14]. However, the aqueous PEDOT:PSS dispersions are at $pH \sim 1$ and corrosive to the ITO

Efficient photovoltaic (PV) cells based on regioregular poly(3-hexylthiophene) (P3HT):fullerene derivative [6,6]-phenyl- C_{61} butyric acid methyl ester (P C_{61} BM) composites have been fabricated by using solution-processed molybdenum oxide (MoO₃) as an anode buffer layer. The influence of solution-processed MoO₃ anode buffer layer on the device performance of P3HT:PCBM PV cells is investigated as compared to the PV cells with different anode buffer layers including water-soluble poly(ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) and the thermally evaporated MoO₃. It is found that the performance of the PV cell with solution-processed MoO₃ anode buffer layer is comparable or higher than the devices using PEDOT:PSS or thermally evaporated MoO₃ as the anode buffer layer. The power conversion efficiency of PV cell with solution-processed MoO₃ anode buffer layer reaches 3.14% under AM1.5G 100 mW/cm² illumination.

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anode that would degrade the long-term stability of the polymer PV cells [15–17]. Metal oxides are good candidates for the anode buffer layers in polymer PV cells. There have been reports suggesting that some metal oxides including nickel oxide (NiO) [18], vanadium oxide (V_2O_5) [15,19] and molybdenum oxide (MoO₃) [15,20–22] are used to replace the PEDOT:PSS as anode buffer layers in polymer PV cells. These reported metal oxides are deposited via pulsed laser deposition or thermal evaporation. In view of the practical large-area production, simple large-scale processing is one of the prerequisites in addition to the high power conversion efficiency (PCE) of polymer PV cells. Therefore, solution-processed anode buffer layers for polymer PV cells are imperative under this consideration.

Herein, we report efficient bulk heterojunction PV cells based on P3HT:PCBM blend by introducing a solution-processed MoO₃ anode buffer layer via spin-coating from aqueous MoO₃ solution. The influence of solution-processed MoO₃ anode buffer layer on device performance of P3HT:PCBM PV cells is investigated as compared to the polymer PV cells without anode buffer (ITO-only) and with PEDOT:PSS or thermally evaporated MoO₃ anode buffer layers. It is found that the PCE of the PV cell with spin-coated MoO₃ anode buffer layer is comparable or higher than the PV cells with PEDOT:PSS and thermally evaporated MoO₃ anode buffer layer.

2. Experimental

The aqueous MoO_3 solution was prepared by hydration method according to the procedure reported by Dong et al. [23]. Ammonium molybdate ((NH₄)₆Mo₇O₂₄) was dissolved in water to

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form 0.01 mol/L solution, marked as solution A. 2 mol/L hydrochloric acid (HCl) water solution was marked as solution B. Solution B was dropped into solution A until the pH value of the mixed solution was adjusted between 1 and 1.5. This mixed solution was marked as solution C, which is aqueous MoO_3 solution.

The aqueous MoO₃ solution was spin-coated onto the substrate and annealed at 160 °C for 10 min in nitrogen-filled glove box for characterization and device fabrication. Fig. 1 shows X-ray diffraction (XRD) patterns of the films fabricated from the aqueous MoO₃ solution and the MoO₃ powders bought from Aldrich Co. Ltd. It can be seen that the film fabricated from the aqueous MoO₃ solution shows the same diffraction peaks as that of MoO₃ powders bought from Aldrich Co. Ltd., which demonstrates a pure phase of the solution-processed MoO₃. The MoO₃ anode buffer layers were prepared by spin-coating the aqueous MoO₃ solution (solution C) onto the ITO glass substrate at 5000, 3000 and 1500 rpm, respectively. Subsequently, the spincoated MoO₃ films were annealed at 160 °C for 10 min in a nitrogen-filled glove box. Fig. 2 shows the scanning electron microscopy (SEM) images of the MoO₃ anode buffer layers spincoated onto the ITO substrate. The MoO₃ nanoparticles are formed on the ITO surface. It is noted that the discontinuous MoO₃ layers are formed and the coverage of MoO₃ nanoparticles on ITO surface decreases with increase in the spin-coating rate.



Fig. 1. X-ray diffraction patterns of the solution-processed MoO_3 and the commercially available MoO_3 powders.

The polymer PV cells have a structure of glass substrate/ITO anode/anode buffer layer/P3HT:PCBM (90 nm)/Al. The polymer PV cells were fabricated on patterned ITO-coated glass substrate with a sheet resistance of 10Ω /square, which had been cleaned by successive ultrasonic treatment in acetone and isopropyl alcohol and then dried at 120 °C for 30 min. The ITO glass was then subjected to UV-ozone treatment for 10 min. Four kinds of polymer PV cells including without anode buffer layer and with PEDOT:PSS, thermally evaporated MoO₃ and solution-processed MoO₃ anode buffer laver are fabricated. For the PV cell with PEDOT:PSS. a 30-nm-thick PEDOT:PSS laver was spin-coated onto the pre-cleaned ITO substrate and baked at 120 °C for 30 min. For the PV cell with thermally evaporated MoO₃, a 5-nm-thick MoO₃ layer was thermally deposited onto the pre-cleaned ITO substrate. For the PV cells with solution-processed MoO₃, the MoO₃ layer was deposited onto the pre-cleaned ITO substrate by spin-coating from the aqueous MoO₃ solution followed by annealing at 160 °C for 10 min. Then, the P3HT:PCBM (1:0.8, 90 nm) active layer was deposited onto anode buffer layers by spin-coating a mixed solution of P3HT (10 mg/mL):PCBM (8 mg/mL) at 900 rpm. Finally the substrates were transferred into an evaporator and pumped down to 4×10^{-4} Pa to deposit 100-nm-thick aluminum (Al) cathodes, producing an active area of 0.12 cm² for each cell. The PV cells were annealed at 140 °C for 2 min on a hot plate inside a nitrogen-filled glove box before electrical characterization. The illuminated current density-voltage (J-V) characteristics of the PV cells were tested using a computer-controlled Keithley 236 source meter under AM1.5G illumination from a calibrated solar simulator. The external quantum efficiency (EQE) of the PV cells was measured with a lock-in amplifier at a chopping frequency of 280 Hz during illumination with the monochromatic light from a xenon lamp.

3. Results and discussion

Fig. 3(a) shows the molecular structures of P3HT and PCBM, which act as electron donor and electron acceptor in the active layer, respectively. Due to the high self-organization capability and high hole mobility of P3HT, the P3HT:PCBM blend film can form nanoscale donor–acceptor interpenetrating networks via thermal annealing or solvent-vapor annealing, which favors exciton dissociation and charge collection [24,25]. The energy level diagram of the P3HT:PCBM blend PV cells with different anode buffer layers is shown in Fig. 3(b). Since the work function of ITO is merely about 4.7 eV, it would result in a low open-circuit voltage (V_{OC}). While the PEDOT:PSS has a work function of 5.2 eV, which would favor improving the V_{OC} of the PV cell. MoO₃ is a



Fig. 2. SEM images of solution-processed MoO₃ morphology on ITO substrate spin-coated from aqueous solution at 5000 (a), 3000 (b) and 1500 rpm (c), respectively.

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