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Improvement in power conversion efficiency and long-term lifetime of organic photovoltaic cells by using bathophenanthroline/molybdenum oxide as compound cathode buffer layer



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ABSTRACT

We demonstrate response and lifetime improvements for organic photovoltaic (OPV) by using organic bathophenanthroline (Bphen) and inorganic molybdenum oxide (MoO₃) as a compound cathode buffer layer (CBL). Not only power conversion efficiency (PCE) increases by 27% but also working and storage lifetimes extend at least 10 and 60 times respectively comparing with PV cell with 5 nm thick Bphen as CBL. The optimized PV structure is ITO/CuPc (20 nm)/C₆₀ (40 nm)/Bphen (2 nm)/MoO₃ (5 nm)/Al and the CBL of the reference device is 5 nm Bphen without MoO₃ layer. The achievement of PV performance and lifetime improvement by introducing such a compound CBL is attributed to appropriate level alignment between C₆₀, Bphen and MoO₃, and stable inorganic MoO₃ can prevent from superior oxygen and moisture diffusion into the C₆₀ acceptor layer. The detailed working mechanism of the compound Bphen/MoO₃ CBL in the OPV cells is also argued.

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1. Introduction

Since Tang's initial discovery of the donor–acceptor organic photovoltaic (OPV) cells based on small molecule planar heterojunction, OPV cells have received increasing attentions in the hope of their application as a renewable, cheaper, and economical power source compared to their inorganic counterparts [1–3]. Significant efforts have been devoted in recent years to improve the power conversion efficiency (PCE); however, there are relatively few studies that develop ways to improve stability of OPV cells, which is also a huge problem restricting the application of OPV cells [4,5]. Therefore, there is a great deal of interest in introducing useful structures into OPV cells to gain both high PV response and long-term lifetime.

Cathode buffer layer (CBL) can play an important role in protecting excitons and holes to reach to cathode surface, thus avoiding quenching of the excitons and holes by the cathode surface states. Besides, CBL can assist in establishing an ohmic

contact between acceptor and cathode [6,7]. The most commonly used CBLs, such as thin bathocuproine (BCP) [8] and bathophenanthroline (Bphen) [9], are intensively used as general exciton blocking layer (EBL). Since wide energy gap semiconductors transport carriers via cathode damage which is induced during metal deposition process, they are unsuitable to be used as thick EBL, which is a disadvantage in improving the working lifetime of OPV cells. Although the defect states induced by cathode deposition are beneficial for electron transport, heat-deposited Al atoms may still damage C₆₀ acceptor. For the stability of OPV cells, it is well known that the predominant degradation mechanism is attributed to the entrance of ambient gases such as oxygen or moisture into the cells, and the major diffusion route of ambient gas is through the top cathode [10,11]. One of the important reasons for OPV cell degradation is the markedly reduced conductivity of C₆₀ film due to the entrance of ambient gases into the active layer. So it is necessary to develop new cell structure to prevent C₆₀ from the damage induced by heat-deposited Al and entrance of ambient gases. On the other hand, it is recognized that MoO₃ has been widely used as anode buffer layer (ABL) to modify indium tin oxide (ITO) either for OLEDs [12,13] or for OPV cells [14–17] due to its hole injection property and increasing work

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function of ITO anode. At the same time, different groups, however, have reported vastly different functions of MoO_3 , such as enhancing electrical conductivity of organic hole-transporting layer [18], preventing an unwanted chemical reaction between ITO and the active layer [19], reducing the electron leakage current [20], forming a Schottky barrier [21] and so on.

In the present article, Bphen/ MoO_3 is used as a compound CBL in OPV cells, in which thinner Bphen can transport electrons by tunneling mechanism and MoO_3 could protect against contamination of Al atoms to C_{60} film and block the moisture and oxygen gas in air to diffuse into OPV cells. Such a CBL is different from traditional single Bphen or BCP because the CBL can integrate strongpoint of organic and inorganic semiconductors. As a result, a maximum PCE of 1.62% for the optimized ITO/CuPc/ C_{60} /Bphen/ MoO_3 /Al cell is obtained, which increases by 27% compared with the reference cell with 5 nm Bphen. More importantly, optimized cell offers enlarged working lifetime. As the PCE of the reference cell is decreased to 0, the improving cell still maintains 73% of the initial PCE under a constant illumination of 50 mW/cm^2 . Besides, the storage lifetime increases at least 60 times, and keeps 60% of initial PCE after the cell is conserved for 2 months in an ambient condition, while the cell without compound CBL degrades very quickly and no PCE can be detected in 24 h. In addition to CuPc-donor OPV cells, we also construct Rubrene donor based cells with the structure ITO/ MoO_3 /Rubrene/ C_{60} /Bphen/ MoO_3 /Al, which can be another forceful evidence for improving device performance by the compound CBL. For optimized Rubrene donor-cell, PCE is improved by 20% comparing with its reference cell, reaching to a PCE of 2.87%.

2. Experiments methods

Patterned ITO coated glass substrates with a sheet resistance of $20 \Omega/\text{sq}$ were routinely cleaned and treated in an ultraviolet-ozone chamber for 20 min immediately before loading into a high vacuum chamber ($\sim 5 \times 10^{-4} \text{ Pa}$). The evaporating rates were kept at $0.5\text{--}1 \text{ \AA/s}$ for organic layers and MoO_3 layer, and $10\text{--}15 \text{ \AA/s}$ for Al cathode. The current–voltage characteristics were measured using a programmable source meter (Keithley 2400) in the dark and under illumination of 100 mW/cm^2 with AM 1.5G simulated solar spectrum from a solar simulator. All measurements were carried out at room temperature and under ambient conditions without encapsulation. We fabricated two series of cells, and they are referred to be as series-1 and series-2; the former's donor is CuPc and donor of the latter cell is Rubrene. More detailed structure will be described in the next section. The PV devices were consecutively illuminated under a 50 mW/cm^2 iodine–tungsten lamp in open circuit state for working lifetime measurements which is corresponding to ISOS-L-1 in Ref. [22]. Storage lifetime measurements were performed in dark under laboratory ambient at room temperature in open circuit state which is corresponding to ISOS-D-1 in Ref. [22].

3. Results and discussion

Fig. 1(a) and (b) depicts schematic structure of two series cells based on CuPc and Rubrene series, respectively. Note that for series-1 MoO_3 ABL is not used, which is attributed to poor effect of MoO_3 anode buffer layer on performance of CuPc cells [15].

Fig. 2(a) and (b) shows the typical current–voltage ($I\text{--}V$) characteristics of improved and corresponding reference cells under 1 sun (100 mW/cm^2) AM 1.5G simulated solar illumination. We can observe that optimized cell structures are ITO/CuPc (20 nm)/ C_{60} (40 nm)/Bphen (2 nm)/ MoO_3 (5 nm)/Al and ITO/

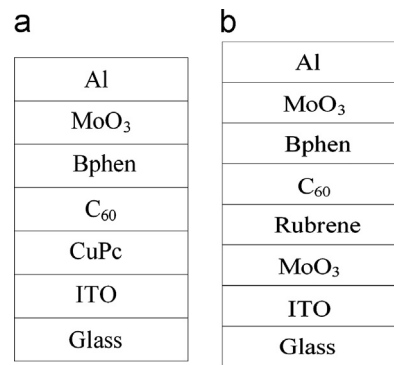


Fig. 1. Schematic structures of two series cells based on (a) CuPc and (b) Rubrene.

MoO_3 (10 nm)/Rubrene (20 nm)/ C_{60} (40 nm)/Bphen (2 nm)/ MoO_3 (5 nm)/Al, respectively, and 5 nm thick Bphen layers are used for both two reference cells. Optimized series-1 and -2 OPV cells offer maximum PCEs of 1.62% and 2.87%, and are respectively enhanced by 27% and 20% comparing with their respective reference cells. In order to understand the working mechanism of Bphen/ MoO_3 CBL, devices with different thicknesses of Bphen and MoO_3 are fabricated. Tables 1 and 2 summarize the PV performances for series-1 and series-2 cells, in which the thicknesses of Bphen and MoO_3 are changed. We can see that 2 nm Bphen plays a very important role in two series improved cells. That is, when 2 nm Bphen layer is not used in CuPc- and Rubrene-OPV cells, PCEs decrease by 60% and 80% for CuPc and Rubrene cells, which are even much lower than those for the respective reference cells. We consider that the poor PV performance can be ascribed to the formation of a new cell of $\text{MoO}_3/\text{C}_{60}$ [23]. This new organic Schottky barrier PV cell provides a reversed PV response, which inevitably decreases the performance of the forward OPV cell. It is also found that when MoO_3 thickness is fixed at 5 nm and the thickness of Bphen increases from 2 nm to 10 nm, the PCE drastically decreases from the maximum to nearly 0. This phenomenon indicates that 5 nm MoO_3 CBL can absolutely prevent Al atom from entering into Bphen layer so that Bphen layer cannot be damaged by heat-deposited Al and damage state cannot be induced. It is impossible for electron transport to occur by tunneling process with a Bphen layer as thick as 10 nm. We also observe that the PCEs decrease when thickness of MoO_3 continues to rise, which is attributed to the increase in cell resistances induced by too thick MoO_3 layer. Thus we select 2 nm Bphen/5 nm MoO_3 to be CBLs for fabricating improved cells. Fig. 3 displays the schematic energy-level diagram for electron transport process in the OPV cell with 2 nm Bphen/5 nm MoO_3 . The LUMO and HOMO levels (units in eV) are cited from Ref. [16] for MoO_3 , Ref. [24] for Bphen, Ref. [25] for CuPc and Ref. [26] for C_{60} . It seems that the electrons can pass through 2 nm Bphen layer efficiently by tunneling mechanism [8,27] and then the electrons transport via LUMO level of MoO_3 reaching to Al cathode as the appropriate level alignment between C_{60} , Bphen and MoO_3 for electron transport. This can be understood as below. Due to the occurrence of interfacial dipole effect, there is a $\sim 3 \text{ eV}$ shift of the vacuum level [28], so that the LUMO level of MoO_3 just lies near the LUMO of C_{60} level and the work function of Al, which could benefit for the electron transport. On the other hand, MoO_3 is an inorganic semiconductor with n-type structure and the majority carriers are electrons, and generally hole-injection at anode side of OPV cells is actually via electron extraction from the HOMO of hole transporter (or donor) through the MoO_3 conduction band to anode [16]. Thus we infer that electron transport should also happen via the LUMO level of MoO_3 .

Not only improvements in above PV performances are achieved but also working lifetime is enlarged by using the compound CBL

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