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Efficiency and stability enhancement of polymer solar cells using multi-stacks of C_{60} /LiF as cathode buffer layer

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ABSTRACT

We report on the improved performance and stability of bulk heterojunction (BHJ) polymer solar cells using five stacks of C_{60}/LiF as cathode buffer layer, which was prepared by alternating deposition of C_{60} and LiF layers. The five-stacked C_{60}/LiF film, even with a large thickness ratio of LiF to C_{60} , exhibits good electrical conductivity. The devices with five stacks of C_{60}/LiF buffer layer show a peak power conversion efficiency (PCE) which is 19% higher than that of the conventional devices with only LiF interlayer, primarily due to the improvement in fill factor (FF) of the device. Moreover, the high efficiency of five-stacked C_{60}/LiF based devices is insensitive to changes in the thickness ratio between C_{60} and LiF layers. Since much thicker LiF can be used in five-stacked C_{60}/LiF film, these devices also show superior air stability as compared to the devices with only LiF interlayer. Therefore, five-stacked C_{60}/LiF is a promising alternative to pristine LiF as a cathode buffer layer in polymer solar cells.

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1. Introduction

Polymer solar cells (PSCs) have attracted much attention in recent years due to their unique advantages of low cost, light weight, and potential use in flexible largearea devices [1–3]. In the past few years, significant progress has been made in this area, and power conversion efficiency (PCE $\equiv \eta$) over 7% has been achieved by several research groups, primarily owing to the development of novel materials and optimization of the active layer morphology [4–7]. However, for practical applications, further improvements in efficiency and stability are required.

In general, bulk heterojunction (BHJ) PSCs with a conventional structure are composed of a photoactive layer sandwiched between an indium tin oxide (ITO) anode and a low-work-function metal cathode. Considering the existence of energy barriers between the active layer and electrodes, anode/cathode buffer layers are used to effectively reduce the charge injection/extraction barriers to allow ohmic contact formation. A widely used cathode buffer layer in PSCs is LiF. Experiments have shown that a thin LiF buffer layer improves not only device efficiency but also device stability in ambient by retarding the oxygen diffusion into organic films [8,9]. However, the thickness of LiF is limited to less than 2 nm due to its insulating property; the thicker LiF will inevitably increase the series resistance (R_s) of the device, which is detrimental to electron collection. Such a thin layer of LiF cannot provide sufficient protection against the diffusion of oxygen and water. In addition, the thin LiF interlayer also cannot efficiently protect the underlying active layer from damage during the evaporation of hot cathode metal atoms. Therefore, it is highly desirable to adopt much thicker buffer layers for better device performance and stability.

Recently, Gao et al. [10] reported the improved efficiency and stability of PSCs using a 30 nm thick C_{60} :LiF (75 wt%) composite as cathode buffer layer, which was deposited by co-evaporation. Nevertheless, the





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co-evaporation method requires in situ and precise control over the deposition rates of each component, and any deposition rate variations will potentially lead to incorrect blend ratios and poor device performance [11]. To address this problem, we introduced five stacks of C₆₀/LiF cathode buffer layer which was prepared by alternating deposition of C₆₀ and LiF via layer-by-layer deposition process. The five-stacked C₆₀/LiF film exhibits good electrical conductivity even when the thickness ratio of C_{60} to LiF is 1:2, namely 2 nm thick C₆₀ and 4 nm thick LiF for one unit $(C_{60}/\text{LiF} \text{ bilayer})$, while the pristine LiF film with thickness larger than 2 nm has a much lower conductivity due to its insulating nature. Using five stacks of C₆₀/LiF as the cathode buffer layer, PSCs based on poly(3-hexylthiophene) (P3HT):[6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) blend show performance that is superior to conventional cells with only LiF buffer layer. The improved device efficiency and stability can be attributed to the better protection of the active layer from hot cathode metal atoms as well as from oxygen and moisture after exposure to ambient air owing to the larger thickness of five-stacked C₆₀/LiF films.

2. Experimental

The schematic device structure of the PSCs studied in this work is shown in Fig. 1. ITO-coated glass substrates (Delta Technologies, LTD) were cleaned in acetone and isopropyl alcohol (IPA) under sonication for 5 min each and then treated by O_2 plasma for 60 s to generate the hydrophilic surface. The filtered poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) solution (H. C. Starck, Clevios PH 500) was spin-coated onto the cleaned ITO electrodes at a speed of 2000 rpm for 50 s, followed by baking at 110 °C for 20 min under nitrogen atmosphere. Subsequently, the samples were transferred to a N₂-purged glovebox for spin-coating of photoactive layer.

P3HT (Rieke Metals Inc., 4002-EE, 91–94% regioregularity) and PCBM (American Dye Source, Purity: >99.5%) were dissolved in chlorobenzene with a weight ratio of 1:1. The mixed solution was filtered using a 0.45 μ m filter, and then spin-coated on top of the PEDOT:PSS layer at 1000 rpm for 50 s, followed by thermal annealing at 130 °C for 20 min, which produced a ~180 nm thick active layer measured using a Dektek surface profiler. The C₆₀, LiF and Al



Fig. 1. Schematic device structure of P3HT:PCBM polymer solar cells using five stacks of C_{60} /LiF as the cathode buffer layer.

(75 nm) electrode were sequentially deposited by thermal evaporation at a base pressure of 1×10^{-6} mbar. The deposition rate and film thickness were monitored with a quartz crystal sensor. A circular-shaped shadow mask of 1 mm diameter was put on the sample to define the active area before the Al deposition. To aid our study, electron-only devices were also fabricated with the structure: ITO/ cesium carbonate (Cs₂CO₃)/P3HT:PCBM/cathode buffer layer/Al. Cs₂CO₃ (0.2 wt%), dissolved in 2-ethoxyethanol, was spin-coated on ITO substrate and subsequently baked at 150 °C for 20 min. For contact resistance measurements, a specially designed shadow mask with varying gap widths between two adjacent strips was used to replace the circular-shaped one during the evaporation of Al cathode.

Current versus voltage (I-V) characteristics were measured using a Keithley 2400 system under simulated Air Mass 1.5 Global (AM 1.5 G) solar illumination at a intensity of 100 mW/cm², which was calibrated by a power meter (OPHIR, Nova-Oriel) and a reference silicon solar cell. The measurements were done with the solar cells inside the glovebox (<0.1 ppm O₂ and H₂O). For stability study, the unencapsulated devices were taken out from the glovebox and stored in the dark under ambient conditions. After a certain time, the same devices were transferred back into the glovebox and subjected to illumination to monitor the decay of the characteristics. The shelf life of solar cells is defined as the time period for the PCE to decay to 50% of its initial value (half-life). Atomic force microscope (AFM) images were taken with a Veeco Dimension-Icon AFM operated in tapping mode.

3. Results and discussion

Fig. 2a shows the current density-voltage (I-V) characteristics, recorded under 100 mW/cm² illumination (AM 1.5 G), of the devices with varying thicknesses of LiF buffer layer. The corresponding photovoltaic parameters are summarized in Table 1. As the thickness of LiF is increased from 1 to 5 nm, both the short-circuit current density (I_{sc}) and fill factor (FF) decrease; together with the almost unchanged open-circuit voltage (V_{oc}), the overall PCE declines gradually. To find the reason for the reduction in both the I_{sc} and FF, we calculated R_s and shunt resistance (R_{sh}) from the slope of photo J-V curve at 0 mA/cm² and 0 V, respectively. The corresponding results are listed in Table 1. It is found that the R_s significantly increases with increasing the thickness of LiF, which explains the drop in J_{sc} and FF. The increased R_s is rationalized in terms of the insulating property of LiF, which is detrimental to the electron collection at the cathode. Additionally, we find that the $R_{\rm sh}$ also increases slightly with the increase of LiF thickness because a thicker LiF layer can provide better protection for the underlying active layer from damage by the energetic metal atoms during the Al deposition.

In comparison with the LiF interlayer, the device with five stacks of C_{60}/LiF as the cathode buffer layer exhibits a much higher efficiency, as shown in Fig. 2b and Table 1. The highest PCE for five-stacked C_{60}/LiF based devices is 3.58% with the J_{sc} , V_{oc} and FF are 9.27 mA/cm², 0.55 V and 70.2%, respectively, while the conventional device

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