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Improved photovoltaic effect of polymer solar cells with nanoscale interfacial layers

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ABSTRACT

In poly(3-hexylthiophene):methanofullerene bulk-heterojunction polymer photovoltaic (PV) cells, we introduced nanoscale interfacial layers between the PV layer and the cathode. The nanoscale interfacial layers were made of ultrathin poly(oxyethylene tridecyl ether) surfactant and low-work-function alloy–metal of Al:Li layers. It was found that the nanoscale interfacial layers increase the photovoltaic performance: increasing short-circuit current density with fill factor and improving device stability. For PV cells with the nanoscale interfacial layers, an increase in power conversion efficiency of $4.18 \pm 0.24\%$ was achieved, compared to that of the control devices ($3.89 \pm 0.08\%$).

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1. Introduction

Recently, important research has been conducted on the development of polymer solar cells (PSCs) for realizing high efficiency in generating electrical power by absorbing light [1–5]. Among the desired developments, an interesting one for the efficient photoinduced charge generation of the photovoltaic (PV) layer is the bulk heterojunction PSC structure [2,3], which is composed of a conjugated polymer and fullerene composite layer.

Using the bulk heterojunction structure together with pre- or post-thermal annealing of PV layers of poly(3-hexylthiophene) (P3HT) and phenyl C_{61}-butyric acid methyl ester (PCBM), PSCs with a power conversion efficiency (PCE) of 3–4% were demonstrated [4,5]. More recently, it was reported that the efficiency can be increased dramatically up to $\sim 6.77\%$ by using a low-band gap polymer cell [6]. The use of these PSC technologies should result in much lower costs for high-speed solution-coating processes, such as the roll-to-roll process (abbreviated R2R) [7,8]. However, for practical applications, the efficiency of PSCs needs to be improved further with good environmental stability. In order to achieve this goal, the electrical contact at the interfaces between the PV layer and the electrodes also has to be optimized. To achieve better electrical contact, it has been common practice to modify the electrodes. At the indium tin oxide (ITO) anode side, a buffer layer of poly(3,4-ethylene dioxylene thiophene):poly(styrene sulfonic acid) (PEDOT:PSS) [9] or self-assembled monolayers [10], has been used to tune the electrical contact of the ITO. At the cathode side, an ultrathin LiF [11] or poly(ethylene oxide) [12] layer has been placed between the PV layer and the cathode to form a favorable dipole layer, which results in improved electron collection. As a representative example, an efficiency of $\sim 3.3\%$ was reported for PSCs with a LiF interfacial layer [11]. Recently, instead of a LiF interfacial layer, a multifunctional inorganic layer of TiO_x [13] or ZnO [14] was provided as a hole-blocking barrier, an optical spacer, and oxygen barrier. The use of these inorganic films resulted in significantly improved device efficiency and stability. A PCE of 4.2% was reported when using ZnO film with the dipolar layer [14]. However, the electrical properties of inorganic films depend strongly on the conditions of processing and their fabrication is complex. These may be limiting factors for fabricating highly efficient devices. Thus, the improvements made so far with respect to the efficient transfer of electrons in PSCs remain inadequate. Hence, to improve PSCs further, we initiated work related to the interfacial layer. In this study, to improve the collection of electrons at the cathode side, we used nanoscale interfacial layers, which consisted of (i) a ultrathin surfactant of poly(oxyethylene tridecyl ether) (PTE) and (ii) a low-work-function metal alloy (Al:Li) layer, as shown in Fig. 1. Because of the low highest-occupied-molecular-orbital (HOMO) (-8.1 eV) and high lowest-unoccupied-molecular-orbital (LUMO) (-2.1 eV) levels of PTE [15], it may be surmised that the nanoscale interfacial layers of (PTE/Al:Li) can modify the charge carrier injection and transport at the electrode interface. In previous work, nanoscale double interfacial layers (DILs) were also used to improve the performance of organic light-emitting diodes [15].

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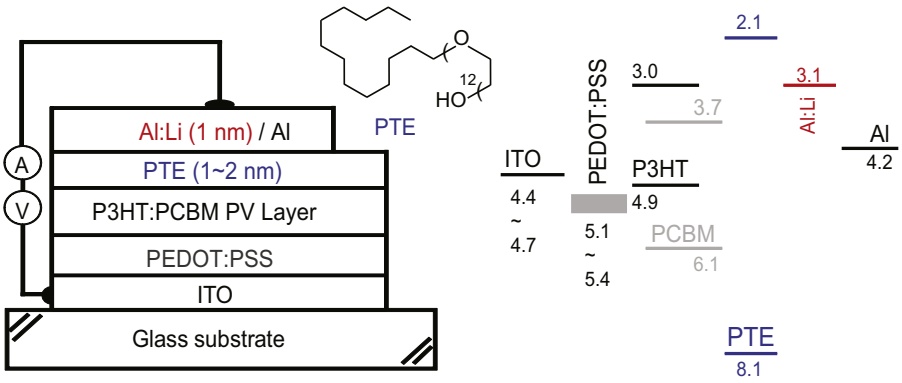

Fig. 1. (Left) Device structure and (right) energy band diagram for the studied polymer solar cells with nanoscale interfacial layers.

2. Experimental details

For the experiments, P3HT (Aldrich, regiospecific ratio $\geq 99.5\%$), as the electron donor and PCBM (Rieke Metals, Inc.), as the electron acceptor were used as received. The chemical structures of P3HT and PCBM are shown in elsewhere [4,5]. P3HT (1.20 wt%) and PCBM (0.88 wt%) were added to a solvent of 1, 2-dichlorobenzene to blend a PV solution [16]. An ITO layer (80 nm, 30 Ω/square) on glass substrate was used as an anode, as shown in Fig. 1. After routine cleaning of the substrate using ultraviolet-ozone treatment, the blended PV solution was spin-coated on top of the ITO, precoated with PEDOT:PSS buffer layers. The PV layer was about 85 nm thick. In order to form the first ultrathin interfacial layer of PTE ($C_{13}H_{27}(OCH_2\ CH_2)_{12}OH$, Aldrich), a PTE solution (0.1 wt% in distilled water) was further spin-coated (1–2 nm) on top of the underlying PV layer. (Details of the thickness of the PTE layer will be reported elsewhere.) After spin-coating, all coated films were baked at 120 °C for 3 min. For the second interfacial layer, a $\sim$1 nm thick Al:Li alloy (Li: 0.1 wt%) layer was formed on the PTE layer via thermal deposition (0.5 nm/s) at a base pressure below 2×10^{-6} Torr. Finally, a pure Al ($\sim$50 nm thick) cathode layer was formed on the interfacial layers under vacuum. Thus, the sample PSC device fabricated has the structure of ITO/PEDOT:PSS/P3HT:PCBM/PTE (1–2 nm)/Al:Li (1 nm)/Al with active area of $3 \times 3\ mm^2$. For comparison, we also fabricated a control device without the PTE interfacial layer, i.e. ITO/PEDOT:PSS/P3HT:PCBM/Al:Li (1 nm)/Al. For post-thermal annealing, the fabricated PSC was annealed at 150 °C for 10 min to induce the crystallization of the PV layer [4,5]. It should be noted that, except for the PTE interfacial layer, the control device was fabricated in exactly the same way as the sample cell. The optical properties of the PV layers were investigated via UV-vis spectrometry at room temperature with a Cary 1E (Varian) UV–vis spectrometer. The performance of the PSCs was measured under an illumination intensity of 100 mW/cm^2 generated by an AM1.5 light source (Newport, 96000 Solar Simulator). The photocurrent–voltage ($J-V$) characteristics were measured with a source meter (Keithley 2400) and calibrated by using a reference cell (Bunkoh-keiki, BS-520). The incident photon-to-current collection efficiency(IPCE) spectra were measured using an IPCE measurement system (Titan Electro-optics Co., QE-IPCE 3000).

3. Results and discussion

First, the optical characteristics of the PV layers were observed by UV–vis absorption spectroscopy. Fig. 2 shows the normalized absorption spectra of PV layers after thermal annealing at 150 °C for 10 min. It is clear from the figure that the P3HT:PCBM PV film

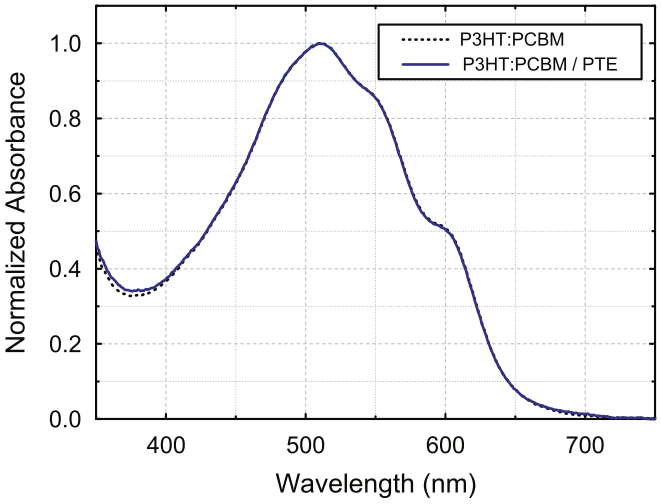

Fig. 2. Normalized UV–vis absorption spectra of PV layers of P3HT:PCBM without (black dotted curve) and with (blue solid curve) a PTE interfacial layer after heating treatment at 150 °C for 10 min.

shows strong absorption spectra with absorption peak centered at around 507 nm with the pronounced three vibronic absorption peaks at ca. 600 nm. This absorption in the visible region is attributed mainly to the P3HT polymer because PCBM shows strong absorption in the UV region. It is noted that the improvement in the ordering of P3HT polymer with thermal annealing can be confirmed from the pronounced three vibronic absorption peaks. As shown in the figure, these results are almost identical to those of the P3HT:PCBM PV film with the PTE interfacial layer. Thus, it indicates that the PTE interfacial layer does not alter the optical absorption characteristics of the PV layer of P3HT:PCBM.

Next, the effect of the interfacial layers on the performance of the current versus voltage ($J-V$) characteristics for the PSCs was investigated, as shown in Fig. 3. Fig. 3(a) shows the dark current characteristics of the PSCs under study. Both of the devices that we tested show an excellent rectification ratio and thus good coverage for the organic layers. However, as shown in the figure, the tested PSCs show a small but clear difference in current flow, which implies that there is a clear difference in the extraction of charge carriers through the interfacial layers. Thus, to see the PV effect of the studied devices, $J-V$ curves under illumination were also observed, as shown in Fig. 3(b). For the case of the control PSC devices without the PTE interfacial layer, fairly good performance was observed with an open-circuit voltage (V_{OC}) of 0.61 ± 0.01 V and a short-circuit current density (J_{SC}) of 11.11 ± 0.17 mA/cm^2

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