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Large area flexible polymer solar cells with high efficiency enabled by imprinted Ag grid and modified buffer layer

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ABSTRACT

To take a full advantage of polymer semiconductors on realization of large-area flexible photovoltaic devices, herein, we fabricate polymer solar cells on the basis of polyethylene terephthalate (PET) with imprinted Ag grid as transparent electrode. The key fabrication procedure is the adoption of a modified PEDOT:PSS (PH1000) solution for spin-coating the buffer layer to form a compact contact with the substrate. In comparison with the devices with intrinsic PEDOT:PSS buffer layer, the advanced devices present a much higher efficiency of 6.51%, even in a large device area of 2.25 cm^2. Subsequent characterizations reveal that such devices show an impressive performance stability as the bending angle is enlarged to 180° and bending time is up to 1000 cycles. Not only providing a general methodology to construct high efficient and flexible polymer solar cells, this paper also involves deep insights on device working mechanism in bending conditions.

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1. Introduction

Polymer solar cells have emerged as a classic paradigm for converting solar energy to electricity, due to their inherent merits including low cost, light weight, flexible texture and rapid energy payback time [1–6]. By using novel polymers with a low bandgap, power conversion efficiencies (PCEs) up to 10% have been reported [7,8]. However, such devices are usually fabricated on ITO based substrates that are relatively expensive and brittle. This hinders the corresponding devices from being manufactured in the industrially preferential roll-to-roll method [9–13]. Additionally, the upcoming huge market for flexible electronics generates enormous demands to flexibilize the according power supplies such as solar cells. To realize high efficient, stable and flexible photovoltaic devices, the brittleness of ITO makes it not applicable for long term bending usage.

So far, nearly all the polymer solar cells with PCE over 8% were fabricated with a small area of less than 0.2 cm^2 [14–16]. With the device area scaling up, the performance of polymer solar cells suffers severe degradation [17,18]. The cumulative resistance of ITO in large area could be responsible for this issue, assuming that the pinholes could be well avoided by optimizing the fabricating procedures. Thus, getting rid of ITO and searching an alternative transparent electrode has attracted extensive attention.

Recently, flexible PET substrates embedded with imprinted Ag-grids have been reported as transparent electrodes for polymer solar cells [19–23], perovskite solar cells [24,25] and organic light-emitting diodes [26]. The high conductive nature of Ag-grid makes the resistance of substrates less susceptible to size enlargement and high transparency could be easily acquired by manipulating the cover ratio of the grid on substrates. Significantly, the high flexibility of Ag line in the grid enables the subsequent devices with good performance stability in multi-cycle bending usage. However, PET is hydrophobic which gives rise to a technical difficulty for spin-coating PEDOT:PSS aqueous solution to form the buffer layer.

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Though PEDOT:PSS has been used in the reported devices, this limit makes the corresponding devices either in a small size or with a relatively low performance [19–21]. A comprehensive work on using the advanced PET/Ag-grid electrode efficaciously is thus still meaningful.

In this paper, with the motivation to develop a convenient technique to fabricate high efficient flexible polymer solar cells in a large scale, we fabricate polymer solar cells with a large area of 2.25 cm^2 on PET/Ag-grid substrates. Such area is nearly two folds of the area in the recording device using similar substrates [22]. By optimizing the structure and component of the devices, including the profiles of Ag-grid and the additives of the buffer layer, a high PCE of 6.51% has been realized. Such efficiency is among the best results for the flexible polymer solar cells in a large area. To be more noteworthy, the devices exhibit good stability against mechanical bending.

2. Materials and methods

2.1. Fabrication of PET/Ag-grid substrates

PET/Ag-grid substrates were fabricated via nano-imprinting technique, the detailed process is described as follows. A photoresist layer with the designed shapes for the Ag-grid (rhombus, square and hexagon) was firstly fabricated by a laser direct writing system. Then the micro-electroforming technique was applied to fabricate the nickel mold with structure which was complementary to the photoresist. The nickel mold was wrapped on the surface of a roller to form a roller mold. A layer of ultraviolet resin with a thickness of 2 μm was coated on the PET as nano-imprint resist. Then roll-to-roll process was applied to fabricate the designed groove grid on the UV resist. The resist was cured under a UV light with a wavelength of 365 nm for 1 h. Then Ag paste was filled into the grooves of the UV resist by a blade-coating process. Subsequently, an infrared-assisted heat cycle oven was used to sinter the Ag paste, forming the conductive Ag-grid network. Finally, a PET protection film was coated on the PET/Ag-grid substrate to keep it clean.

2.2. Fabrication of flexible solar cells

Electron donor material PTB7-Th and cathode interface material poly [(9,9-bis(3′-(N,N-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9–dioctylfluorene)] (PFN) were purchased from 1-material Inc.. Electron acceptor material $PC_{71}BM$ was purchased from Sigma-Aldrich. PEDOT:PSS (Clevios PH1000) was obtained from H.C. Starck Clevios. Fluorosurfactant Capstone® FS-31 was supplied by Dupont. All the materials and chemicals were used as received. The PET/ITO substrate was supplied by Zhuhai Kaivo Optoelectronic Technology Co., Ltd.

Fabrication procedures of the flexible solar cells are as follows. PET protection film was peeled off from the PET/Ag-grid substrate, and substrates were treated with UV-ozone for 10 min, then PEDOT:PSS (or PEDOT:PSS +5% DMSO+0.1% FS-31, by volume) solution was spin-coated on the substrates at 1000 rpm for 60 s and baked at 120 °C for 30 min. Next, Zn precursor was spin-coated on the substrates, following by annealing in air at 150 °C for 30 min. The Zn precursor was prepared by dissolving zinc acetate dehydrate (1 g) and ethanolamine (0.28 g) in 2-methoxyethanol (10 mL) under vigorous stirring for 12 h for the hydrolysis reaction in air [27]. After that, the substrates were transferred into nitrogen glove box and a thin PFN layer was spin-coated on the substrates. Then, blend films of PTB7-Th: $PC_{71}BM$ (with concentration of 10 mg mL^{-1}: 15 mg mL^{-1}) were cast from a mixed solvent of o-dichlorobenzene and 1,8-diiodoctane (97:3, vol%) at 900 rpm for 2 min, forming the active layer. Finally, 5 nm MoO_3 and 150 nm Ag were deposited by thermal evaporation as anode. As to the PET/ITO based devices, fabrication procedure is similar, only without the PEDOT:PSS layer.

A schematic fabrication flow chart is given in the supporting information (Fig. S1).

2.3. Characterization

Transmittance measurement was conducted with ultraviolet–visible spectrophotometer (UV–vis, Persee TU-1950). The cross-sectional SEM image of the device was measured by JSM-7401F. The J-V characteristics were obtained using a Keithley 2450 source measure unit under AM 1.5G illumination with an intensity of 100 mW/cm^2. IPCE measurements were performed at QEPVSI-B Measurement System (Newport). Impedance spectroscopy (IS) measurement was conducted with a CHI electrochemical workstation under dark conditions, with an oscillating voltage of 10 mV and frequency scanning range of 10 Hz to 1 M Hz. The solar cells were held at the respective open circuit voltage obtained from the J-V curves while the IS spectrum was recorded. Atomic force microscopy (AFM) images were taken on Bruker Dimension Edge.

3. Results and discussion

The PET/Ag-grid substrates were prepared by filling Ag paste into the highly ordered concave mesh in PET substrates. The resulting embedded Ag-grid is superior over the convex counterparts by effectively reducing the possibility of pinhole generation. Importantly, the procedure was performed in a large scale roll-to-roll manner, thus beneficial for industrialization of the devices. As shown in Fig. 1a, the flexible and transparent substrates could be fabricated with width around 50 cm and unlimited length. The patterns of the Ag-grid are tunable depending on the molds for imprinting the concave mesh. More details in the advantages of the used PET/Ag-grid substrates are given in the Supporting Information (Section 1). Herein, we selected three patterns for comparison: rhombus, square and hexagon, with side length of 440 μm, 220 μm and 200 μm, respectively. The corresponding transmittance spectra are given in Fig. 1b and the substrates with these patterns present a good and stable transmittance in the region of 400–800 nm, while the traditional PET/ITO substrate shows an obvious transmittance decrease with wavelength below 600 nm. The plateau transmittance values of these PET/Ag-grids are around 90%, higher than that of PET/ITO. These data demonstrate the superiority of PET/Ag-grid substrates over the traditional PET/ITO counterpart. The similarity of the transmittances of different patterned PET/Ag-grids arises from the similar coverages of Ag on PET.

Fig. 2a shows the schematic architecture of the corresponding flexible solar cells constructed by PET/Ag-grid/PEDOT:PSS/ZnO/PFN/PTB7-Th:$PC_{71}BM$/MoO_3/Ag. The according cross-sectional scanning electron microscope (SEM) image is given in Fig. 2b, where the embedded Ag-grid is about 2.8 μm in depth and has a compact contact with PET and the subsequent componential layers of the devices. For real applications, the devices were fabricated in a large area of 2.25 cm^2, as shown in Fig. S2. Energy levels of each layer are illustrated in Fig. 2c and the alignment is preferential for the conducting excited electrons to Ag-grid/PEDOT:PSS and the collection of holes by Ag.

Attributing to the insulating nature of PET, it is necessary to prepare a thin layer of conducting polymer, PEDOT:PSS on PET/Ag-grid substrate to improve the charge collection efficiency. The hydrophobic nature of PET, however, makes it hard to spin-coat PEDOT:PSS aqueous solution. As shown in the inset of Fig. 3a, the poor wettability of PEDOT:PSS solution on PET renders an inferior spread of the solution. To address such dilemma, two additives

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