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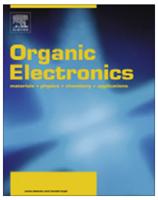

Unexpected solid–solid intermixing in a bilayer of poly(3-hexylthiophene) and [6,6]-phenyl C61-butyric acidmethyl ester via stamping transfer

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ABSTRACT

The transferred bilayer active layer (P3HT/PCBM) prepared from a stamping transfer technique can be expected to undergo a sharp increase in the concentration profile at the P3HT/PCBM interface. However, the PCBM top layer is unexpectedly well blended with the P3HT bottom layer during the solid film stamping process with the help of the residual solvent. Auger spectroscopy confirmed that the bilayer have almost same composition from the anode side to the cathode side as the bulk-hetero junction film. This unique discovery can be applied in preparing concentration-graded BHJ film from controlling solid–solid intermixing behavior.

1. Introduction

Photovoltaic (PV) devices based on polymer–fullerene thin films have many powerful advantages, particularly with regard to flexible devices, capability with large-area optoelectronics, and simple fabrication from solution processability [1–9]. In the case of bilayer systems, most generated electron–hole charge carriers can be easily recombined and trapped in an active layer before power is generated from the charges because organic semiconductors have a short exciton diffusion length in the scale of 1–10 nm [10,11]. Accordingly, a donor–acceptor blending system of a bulk-hetero junction (BHJ) structure has been researched as a promising alternative for enhancing device performance. The enhancement is due to the large interfacial contact area, therefore the BHJ structure overcomes the low charge carrier mobility. However, the bilayer device efficiency also can be enhanced when several factors that we elaborated in a recent paper (namely effective nano-pattern structures [12,13], compositional control of donor–acceptor materials [14,15], and a concentration-graded intermixed zone) are incorporated into a double active layer system [16,17].

Generally, many of the research groups use spin-coating process for small sized cell or roll-to-roll process for large scale organic PVs to introduce active layer [18–21]. However, the fabrication of a bilayered active layer in PV device is constrained by the common solubility of p-type and n-type materials in most organic solvents. To overcome this drawback, several researchers recently reported the use of a stamping transfer technique to fabricate organic PVs [21,22].

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The transferred bilayer active layer pure poly(3-hexylthiophene)/pure [6,6]-phenyl C61-butyric acidmethyl ester (pure P3HT/pure PCBM) should have discrete composition profile at the P3HT/PCBM interface. This study demonstrates, that a bilayer of P3HT and PCBM prepared by means of a stamping transfer method has an unexpected film composition that is, similar to the composition gradient of BHJ film generated from a P3HT/PCBM blended solution. In this study, we also discuss how a intermixing of P3HT/PCBM bilayer is occurred and affects the performance of the device.

2. Experimental details

First, a ITO glass was cleaned by chloroform, acetone and isopropanol to remove residual organic materials on the ITO surface which was then treated by oxygen plasma for 10 min in order to enhancing hydrophilic property. A sample of poly(3,4-ethylene dioxythiophene: poly(styrene sulfonate) (PEDOT:PSS, Baytron P), which is conducting polymer of hole transporting material was diluted to methanol and spin-coated to a thickness of approximately 35 nm. Using a digital-controlled hotplate, we pre-baked the devices in PEDOT:PSS at 200 °C for 5 min to evaporate the water contents. The stamping-transferred bilayer device, which composed of P3HT (approximately 150 nm) and PCBM (approximately 85 nm) from chlorobenzene, was well coated on the UV-PC film (as shown in inset of Fig. 1 and Fig. S1). Each P3HT and PCBM layer was then individually transferred from the UV-PC film to the PEDOT:PSS-coated ITO substrate, which was heated on a hotplate at 90 °C. The UV-PC film with the P3HT or PCBM layer was attached to the heated substrate and rubbed with cotton swabs with uniform force of 18.8 kg/cm^2 until the P3HT or PCBM layer coated on the PC film totally adhered to the heated substrate. When the stamping transfer process was conducted with simple physical force at a suitable temperature, the coated layer from the UV-PC film was perfectly transferred to the PEDOT:PSS-coated ITO with a reproducibility. A TiO_x interlayer with a thickness of approximately 3 nm was inserted between the bilayer and the Al cathode to enhance the device performance [3]. Subsequently, an Al metal electrode with a thickness of 150 nm was thermally deposited under a pressure of 1.7×10^{-6} Torr. To improve the interfacial contact between the active layer and the Al cathode, we used a thermal annealing process that involved the use of a digitally controlled hotplate at 150 °C for 30 min in a glove box. The active area of the printed bilayer with the thermally deposited Al electrode was correctly confirmed in the range of 3–6 mm^2 with a video microscope (SV-35). The *J–V* curves were measured at an illumination rating of AM 1.5 by using a Keithley 2400 source measure unit. A 1000 W xenon lamp of Oriel 91193 which is intensity was calibrated using a silicon reference cell (Fraunhofer ISE, Certificate No. C-ISE269) served as the light source to the devices. The surface morphology and the roughness of the transferred bilayer were analyzed with an atomic

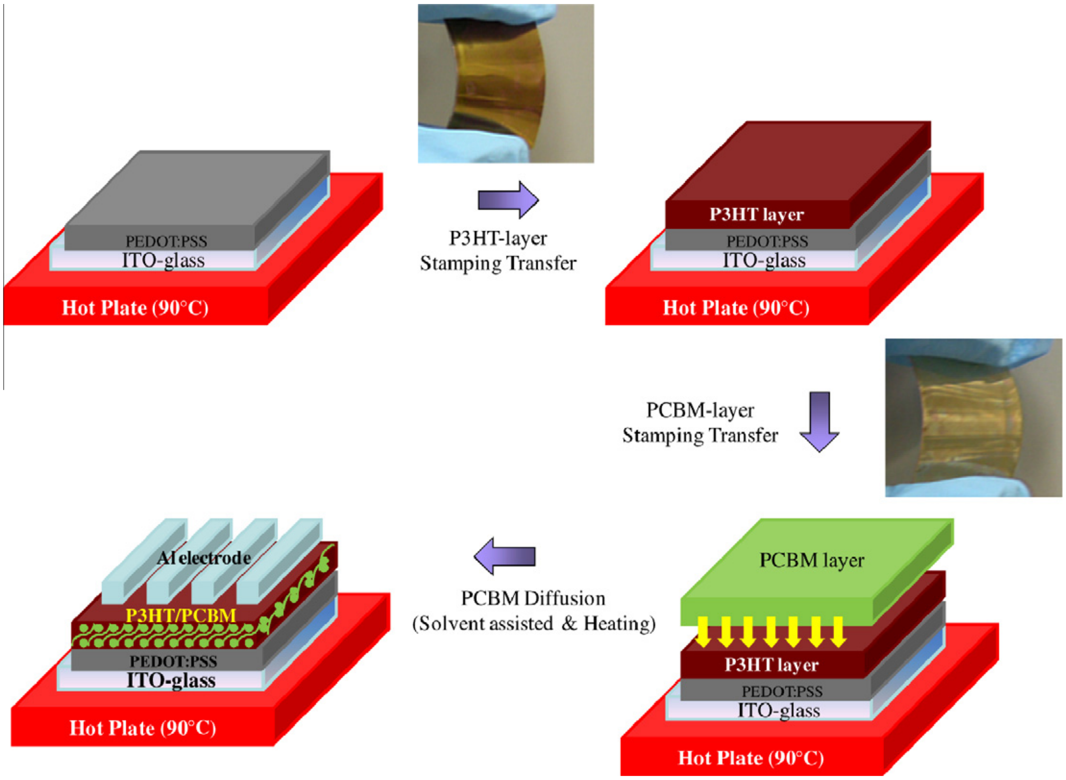

Fig. 1. Schematic diagram of the proposed device structure produced by using a stamping transfer technique of the P3HT/PCBM bilayer with a UV-curable resin-coated polycarbonate film.