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P3HT based solution-processed pseudo bi-layer organic solar cell with enhanced performance

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ABSTRACT

We present a solution-processed pseudo bi-layer organic solar cell with poly(3-hexyl thiophene) (P3HT) as donor and indene-C_{60} bisadduct (ICBA) as acceptor. The devices were fabricated by sequential processing of the active components followed by a thermal annealing treatment. An efficiency of 5.9% was achieved under AM 1.5G irradiation (1000 W/m^2). The obtained efficiency is attributed to an enhanced nanomorphology that arises from the inter-diffusion of the ICBA molecules into a layer of pre-organised polymer (P3HT) and also due to the subsequent crystallisation of the ICBA molecules. These processes facilitate efficient charge generation and extraction. Time of flight-secondary ion mass spectroscopy (TOF-SIMS) depth profiling was carried out for different thermal annealing treatments of these pseudo bi-layer devices, which reveals full inter-diffusion of ICBA into the polymer P3HT. Photo-CELIV (charge extraction by linearly increasing voltage) studies elucidates that the thermal annealing imparts crystallinity to the fullerene phase which results in the improvement of charge carrier mobility.

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1. Introduction

There are mainly two manufacturing protocols that are used for the fabrication of organic solar cells – one is a solution-processed coating procedure and the other is vacuum deposition based manufacturing technique. Each of the above mentioned techniques has their own advantages. The efficiency is steadily on the rise in the last decade and as of now an impressive efficiency value of around 8–9% is achieved for the solution processed bulk heterojunction devices [1–3] and an efficiency of 9.8% has been achieved for vacuum deposited organic solar cells based on small molecules [4]. The increase in the efficiency is attributed to various factors such as (a) design of new polymers and small molecules with an optimum band gap for better utilisation of the photons from the solar spectrum (b) new donor: acceptor combinations (c) understanding the basic device physics that governs the organic solar cell operation and (d) innovative approaches to optimise the microstructure of the blend [5,6]. A key advance in the development of such organic photovoltaic devices was the introduction of the bulk heterojunction (BHJ) concept [7,8]. In this bulk heterojunction concept the donor and acceptor phases were intimately mixed on a nanometre scale but at the same time it brings along difficulties in controlling the nanomorphology, which is crucial for the device performance and often makes reproducibility a major concern [9,10]. Another way of fabricating organic solar cells is the bi-layer heterojunction methodology [11]. This concept was not successful in yielding high efficiency values initially [12,13]. Ayzner et al. recently revisited this concept and fabricated P3HT:PCBM bi-layer solar cells having efficiency values around 3.5% using orthogonal solvent approach [14]. Taking cue from this interesting result few other research groups have also fabricated bi-layer organic solar cells with increasing efficiency values [15–19].

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Recently, we have reported a novel pseudo bi-layer organic solar cell using the standard active materials, poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl C61-butyric acid methyl ester (PCBM) [20]. Our process methodology involved the spin coating of two individual layers of the active materials – P3HT as bottom layer that was spin casted using ortho-dichlorobenzene and the top layer being PCBM that was spin casted using dichloromethane which serves as an orthogonal solvent. Rapid thermal annealing assists in the inter-diffusion of the fullerene molecules from the top layer into the preorganised P3HT bottom layer creating an intermixed donor: acceptor system. TOF-SIMS analysis and X-ray photoelectron spectroscopy (XPS) studies have been used to investigate the nanomorphology of the inter-diffused pseudo bi-layer structure and the results demonstrated that the above mentioned sequential deposition process yielded higher efficiency values than a comparable conventional bulk heterojunction.

This present paper is focused on fabricating pseudo bi-layer organic solar cells using sequential processing technique with ICBA as the electron acceptor and P3HT as the electron donor. The efficiency of P3HT:PCBM based solar cells is limited due to the following factors (a) limited light harvesting by the polymer P3HT due to a relatively high band gap ($\sim$1.9 eV) and (b) low open circuit voltage (V_{OC}) of approximately 0.6 V that is attributed to the small energy difference between the highest occupied molecular orbital (HOMO) of P3HT and the lowest unoccupied molecular orbital (LUMO) of PCBM [21]. A novel indene-C60 bisadduct (ICBA) was synthesised recently by Li et al. with a higher LUMO energy level of -3.91 eV which is 0.17 eV higher than that of PCBM. ICBA was used in fabricating solar cells based on P3HT as electron donor which showed a high V_{OC} of 0.84 V [22]. We explored the potential of this new fullerene material in the pseudo bi-layer configuration and carried out detailed optimisation studies of the solar cells based on P3HT:ICBA. Even though the devices were prepared by sequential processing technique using orthogonal solvents, a strong inter-mixing of the active components was observed due to the inter-diffusion of the ICBA molecules into the P3HT layer by thermal annealing. The inter-diffusion process in these pseudo bi-layer P3HT:ICBA cells were studied using Time of flight-secondary ion mass spectrometry (TOF-SIMS) depth profiling measurements. Thermal annealing treatments (variation of annealing temperature and annealing time) were undertaken to study the morphology evolution of these devices. The relatively high performance of these solar cells is attributed to the strong absorption, effective exciton dissociation as revealed from the photoluminescence quenching and effective extraction of charge carriers as evidenced from the j–V curves, EQE (external quantum efficiency) and Photo-CELIV (charge extraction by linearly increasing voltage) measurements.

2. Materials

The materials used in the present investigation were as follows: regio-regular poly(3-hexylthiophene) ($\geqslant$98%, product number 698997, average M_w 54,000–75,000) was purchased from Sigma–Aldrich. Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS – Heraeus Clevios P VP AI 4083) was purchased from Ossila limited. Indene-C60 bisadduct (ICBA, product code LT-9030) was purchased from Luminescence Technology Corporation. All the solvents that were used in the present investigation were purchased from Sigma–Aldrich. Aluminium (Al) was purchased from STREM chemicals Inc. and Calcium (Ca) was purchased from K.J. Kurt Lesker & Co. All the above mentioned materials were used as received. The molecular structure of P3HT and ICBA is shown in Fig. 1a.

2.1. Experimental

2.1.1. Device fabrication

Pseudo bi-layer organic solar cells based on P3HT:ICBA were prepared on pre-patterned indium tin oxide (ITO, Xinyan Technology Ltd) substrates. The sheet resistance of the ITO (thickness: 90 ± 10 nm) was around 15–20 Ω/s Firstly, the ITO substrates were thoroughly cleaned using a detergent solution under flowing tap water. Successive sonication treatment in deionised water, acetone and isopropanol for 15 min each was then carried out and the substrates were then dried in an oven for 3 h. This was followed by UV-ozone treatment for 15 min and then the PEDOT:PSS solution was filtered through a 0.45 μm cellulose filter and was later spin-coated at 5000 rpm for 60 s to obtain an approximately 50 nm thin layer. The PEDOT:PSS coated substrates were annealed in nitrogen atmosphere for 20 min at 140 °C. Regioregular P3HT in ortho-dichlorobenzene (ODCB) at a concentration of 22.5 mg/ml was prepared and the solution was stirred and heated at 60 °C overnight inside a glove box (Charslton Technologies, <1 ppm moisture and O_2) before being cooled to room temperature and spin cast onto the PEDOT:PSS-coated ITO substrates at 2200 rpm for 80 s. The as spun P3HT films were then allowed to dry in the N_2 atmosphere for at least 20 min prior to the spin coating of the ICBA over layer. ICBA solution was prepared in dichloromethane (DCM) solvent at a concentration of 7.4 mg/ml. The solution was stirred at room temperature for an hour to ensure maximal solubility. Later the ICBA solution was spin cast at 3000 rpm for 10 s onto the P3HT films after a waiting period of 30 s (from the time the ICBA solution was dropped onto the P3HT layer). Then the ICBA coated P3HT film was annealed at either 140 °C or 150 °C for specific amount of time. The optimised device's active layer (P3HT + ICBA only) thickness was found to be around 120 nm. The device was then completed by evaporating a metal cathode consisting of 30 nm Ca/80 nm Al through a shadow mask, resulting in an active device with an area of 9 mm^2. Each ITO substrate contained 6 devices. For the bulk heterojunction solar cells, P3HT and ICBA were dissolved together in 1 ml of o-dichlorobenzene. The concentration of P3HT was 23 mg/ml, whereas the ICBA concentration was found to give best results at 25.3 mg/ml. The optimum spin speed was found to be 800 rpm. Thermal annealing was done at 150 °C for 10 min prior to the deposition of the metal electrodes.

2.1.2. Instrumentation

For the TOF-SIMS measurements, P3HT and ICBA layers were spin coated onto a glass substrate in a similar fashion to that of the solar cell fabrication. Sulphur is used as the

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