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P₃HT:PC₆₁BM based solar cells employing solution processed copper iodide as the hole transport layer

Sayantan Das^{a,b}, Jea-Young Choi^b, T.L. Alford^{a,b,*}^a Department of Chemistry and Biochemistry, Arizona State University, Tempe, AZ 85287, USA^b School for Engineering of Matter, Transport and Energy, Arizona State University, Tempe, AZ 85287, USA

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ABSTRACT

A solution based approach to deposit a p-type CuI hole-transport layer that replaces PEDOT:PSS layer in the fabrication of high-efficiency poly(3-hexylthiophene):[6,6]-phenyl C₆₁-butyric acid methyl ester (P3HT:PCBM) solar cells is reported here. X-ray diffraction analysis identifies the cubic γ -phase of CuI. A Kelvin probe measurement technique is utilized to identify the effective work function of CuI coated ITO. The device optimization is done by varying the concentration of CuI in the precursor solution which played an important role in the efficiency of the solar cell devices. In an effort to explore new inexpensive hole conducting materials for organic solar cells, we have identified copper iodide as a possible alternative. Moreover, the low temperatures required to process CuI films make it a perfect candidate to be used in organic solar cells on flexible substrates.

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1. Introduction

The relentless increase in energy demand in today's world has led to the search for newer renewable energy sources and photovoltaics appeared to be one of the best alternatives for energy production. Ever since organic solar cells (OSCs) gained a reputation of the successful conversion of solar energy to useful electrical energy, numerous efforts have been made to increase the power conversion efficiencies (PCE) by development of new photoactive materials, electron and hole transporting medium; as well as, designing a new transparent electrode system to replace the expensive indium tin oxide [1–12]. Moreover, organic solar cells present many advantages; including the use of low-cost, light-weight materials, compatibility with flexible substrates, low-temperature and roll-to-roll manufacturing techniques [13–15]. The most widely researched photoactive layer to date is a blend of regioregular poly(3-hexylthiophene)(P3HT) and the fullerene derivative [6,6]-phenyl-C₆₁ butyric acid methyl ester (PC₆₁BM) [16]. Under illumination, the blend of the two materials (P3HT:PC₆₁BM) generates excitons which are then efficiently segregated to charge carriers at the interface of the bulk-heterojunction (BHJ) nanostructure and carried towards respective electrodes.

In case of organic solar cell devices, the efficient charge transport through active layers towards the respective electrodes is extremely essential to minimize charge recombination which results in low

efficiency in typical applications. However, the search for a suitable interfacial layers between electrodes and active layer still remains. Hole transport layers (HTLs) like poly(3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT:PSS) are widely used in BHJ cells to ensure Ohmic contacts, efficient hole collections and ITO planarization [17,18]. At the same time, the acidic nature of PEDOT:PSS makes the indium tin oxide (ITO) surface vulnerable to degradation at elevated temperatures [19,20]. Several metal oxides have been demonstrated as efficient hole transport layers in organic solar cell devices. However, the deposition of these oxides generally involves evaporation of the materials in high vacuum tools, which are considerably more expensive and complicated than solution-based processes [14,21–23].

Cuprous iodide (CuI) has a wide band gap (3.1 eV) and has three crystalline phases [24,25]. Amongst the different phases, CuI in the γ -phase with zinc-blende structure has p-type character. Zhou et al. first reported the use of CuI layer in ZnPc:C₆₀ based bilayer organic solar cells, where the CuI nanocrystals are evaporated onto an ITO surface at a glancing angle and is followed by deposition of ZnPc [26]. The investigators suggest that the strong interaction of ZnPc and CuI enables the formation of ZnPc nanopillar arrays. The pillar structure helps to improve light absorption and increases surface roughness induced exciton dissociation. Shao et al. showed that thermally evaporated CuI not only provides Ohmic contact with the active layer (P3HT:PC₆₁BM), but also results in vertical orientation of π - π stacking planes of P3HT with respect to the substrate [27]. They show that the hole mobility of P3HT:PC₆₁BM blend film deposited on CuI surface is higher when compared to the films deposited on PEDOT:PSS because the vertical π - π stacking planes of P3HT induces efficient CuI hole collection in polymer photovoltaic cells. In this

* Corresponding author at: Department of Chemistry and Biochemistry, Arizona State University, Tempe, AZ 85287, USA. Tel.: +1 480 965 7471.

E-mail address: TA@asu.edu (T.L. Alford).

work we show a solution processed CuI film as an effective HTL in P3HT:PC₆₁BM solar cell. It is found that the concentration of CuI played an important role in determining the device efficiency. This work implies that the inexpensive and solution processed CuI can effectively curtail the manufacturing time of low-cost organic solar cell devices in roll-to-roll fabrication.

2. Experimental details

2.1. Reagents and materials

The PEDOT:PSS and the electron donor material regioregular poly (3-hexylthiophene) (P3HT) materials were purchased from domestic chemical vendors. The electron acceptor material [6,6]-phenyl C61 butyric acid methyl ester (PC₆₁BM), copper iodide (CuI) and 1,2-dichlorobenzene were obtained Sigma Aldrich. All these commercially available chemicals were used as-received without further purification.

2.2. Device fabrication

All devices in this work were prepared on 40 Ω sq⁻¹ ITO coated glass substrates. The substrates were cleaned in sequential ultrasonic baths of acetone, methanol, and isopropanol, followed by ultraviolet ozone (UVO) treatment for 10 min. Electron donor material P3HT and electron acceptor material PC₆₁BM were weighed (1:0.8 w/w) and dissolved in 1,2-dichlorobenzene (DCB) in a nitrogen-filled glove box. The solution was stirred for 12 h at room temperature. The PEDOT:PSS solution was spin-coated at 5000 rpm for 60 s on the cleaned ITO substrates, followed by baking at 150 °C for 15 min on a hot plate resulting to a thickness of 30 nm. Different amounts of CuI were weighed out and dissolved in acetonitrile solvent. This solution was used as the precursor solution for spin coating the CuI layer. The different concentrations of CuI solutions were then spin coated on cleaned ITO substrates at 4000 rpm for 60 s, followed by baking on a hot plate at 80 °C inside the glove box. The active layer materials were spin-coated onto the freshly prepared electrodes. Photoactive layers were spin-coated from P3HT:PC₆₁BM blends at 600 rpm for 1 min, and then annealed for 30 min at 120 °C on a hot plate inside the glove box. This corresponded to a layer thickness of 250 nm. Finally, all device fabrication was completed by thermally depositing 15 nm BCP followed by 80 nm Al cathode deposition vacuum (~10⁻⁶ Torr). The thin BCP film acted as an exciton or hole-blocking layer (EBL or HBL). A shadow mask was used during thermal evaporation to define an active area of 0.2 cm². The completed device was then transferred to the solar simulator for testing.

2.3. Thin film characterization and device testing

The optical transmittance of PEDOT:PSS and CuI thin films was measured using a Ocean Optics double channel spectrometer (model DS200) in a wavelength range of 300–800 nm. The structural properties of the as-deposited and annealed CuI films were investigated by X-ray diffraction (XRD) on a Panalytical X'PertPro X-ray diffractometer (XRD) with monochromated Cu Kα irradiation (λ = 1.5418 Å). Surface morphology of the CuI layers on ITO obtained from different concentrations of precursor solution was acquired by using atomic force microscopy (AFM). Work function of CuI layers was measured with a Kelvin probe system (model KP-6500) under inert nitrogen gas atmosphere. A highly oriented pyrolytic graphite (HOPG) was used for reference work function which is known to be 4.6 eV. Current density–voltage (J–V) measurements were performed under simulated AM 1.5 global solar irradiation (100 mW/cm²) using a xenon-lamp solar simulator (Spectra Physics, Oriel Instruments, USA). The light source was calibrated with a standard Si photodiode reference cell

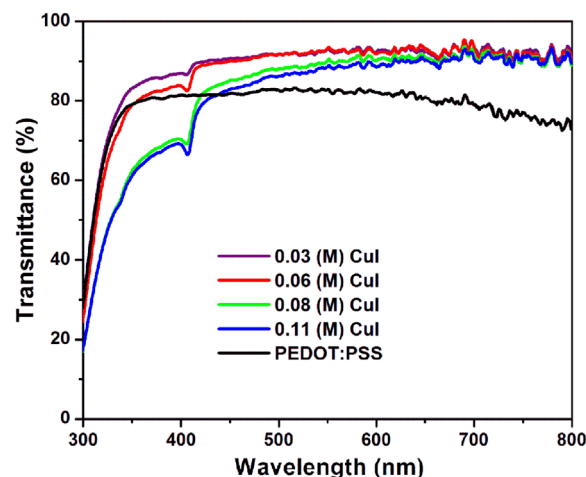


Fig. 1. Optical transmission spectra of PEDOT:PSS and thin films of CuI on glass substrates.

(Hamamatsu Photonics, Japan) prior to measurement. The device external quantum efficiency (EQE) as a function of wavelength was determined from the photocurrent generated in the device from an incident monochromatic light source using an Optronic Lab OL750 series spectroradiometer.

3. Results and discussion

Fig. 1 illustrates the transmission spectra of PEDOT:PSS film and thin films of CuI on glass substrates. It reveals that all of these films were highly transparent in the visible region between 400 and 800 nm. Moreover, all the CuI films exhibit higher transparency than the PEDOT:PSS films in the region between 500 and 800 nm. Interestingly, a hump is observed at about 407 nm and this is due to the excitation of electrons from the subband in the valence bands to the conduction bands of CuI [28]. The size of this hump increases with increasing the amount of CuI in the films. With the increase in the concentration of the CuI precursor solution the transmittance decreases due to increase in thickness of the CuI layer.

Fig. 2 shows XRD patterns regarding the structure of as-deposited and annealed CuI thin films on glass substrate. The spin coating and annealing of CuI films are done inside a glove in order prevent the oxidation of CuI to copper oxide. The JCPDS card 83-1137 is used to identify the cubic γ-phase of CuI [25]. The CuI peak at 25.5° (2θ) corresponds to the (111) reflection of the cubic structure of copper iodide. The as deposited CuI films show broad (111) peak which indicates that the film consists of nanometer sized grains. After annealing for 10 min at 80 °C, the full width at half-maximum (FWHM) of the CuI (111) peak decreases slightly which indicates grain growth. Moreover, a considerable increase in the intensity of the CuI (111) peak is observed and another weak peak at 52.3° (2θ) corresponding to (222) is found which also indicates that the crystallinity of the CuI film is improved after the annealing process at 80 °C.

The device structure of the OSCs and the schematic energy diagram of the materials in devices are illustrated in Fig. 3. The energy level diagram in Fig. 4 suggests that CuI represents a suitable hole transport layer in P3HT:PC₆₁BM type BHJ solar cells. The morphology of the interfacial layers plays an important role in both charge collection and transport in organic photovoltaic devices. Fig. 5 shows 2 × 2 μm² tapping mode AFM images of the different CuI layers on ITO. Greater surface roughness of a layer will impede formation of an intimate contact with the active layer which results in a reduced charge transport efficiency at the interface, as a consequence, decrease the device performance. The surface root-

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