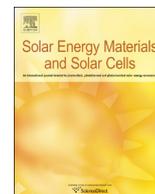




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Enhanced stability in polymer solar cells by controlling the electrode work function via modification of indium tin oxide

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ABSTRACT

To enhance the work function of indium tin oxide (ITO) and make its surface hydrophobic, triethoxy (1H,1H,2H,2H-perfluoro-1-octyl)silane (FOTES) was introduced as a self-assembly monolayer (SAM) material. A polymer solar cell has been fabricated with FOTES–ITO, and due to decreased series resistance (R_s) and increased shunt resistance (R_{sh}), the short circuit current density (J_{sc}), open circuit voltage (V_{oc}), and fill factor (FF) of the optimized device were 9.2 mA/cm², 0.63 V and 57.9%, respectively. The calculated power conversion efficiency (PCE) was 3.4%. In addition, the air stability of the fabricated device was improved.

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1. Introduction

The renewable, light-weight, and low-cost polymer-fullerene bulk heterojunction solar cells (BHJ PSCs) have received great attention for next-generation solar cells [1–5]. Spin-coating, ink-jet printing, roll-to-roll printing, brush painting, or stamping techniques can be applied to flexible substrates to inexpensively fabricate light-weight, flexible, large-area photovoltaic devices [6–11]. However, at 8–9%, the power conversion efficiency (PCE) of PSCs remains too low for commercialization [12,13]. To enhance the PCE in PSCs, many studies have been conducted, and have involved the synthesis of new narrow-band-gap materials for improving the photon harvesting properties, optimization of the morphology in bulk heterojunction (BHJ) films, interfacial modification for better charge carrier collection, and design of novel device structures [14–17].

Another critical factor for commercialization is improvement of the stability. Many of the studies on how to increase life time have focused on work function control, and the improvement of stability through the modification of the anode. Poly(3,4-ethylene dioxathiophene)/poly(styrene sulfonate) (PEDOT:PSS) is one of the most widely used anode buffer materials in PSCs. It has advantages of easy handling, high transparency, and good conductivity, because it enables the usage of solution processing. In addition, the high work function (5.0 eV or above) is an improvement over the low work function of ITO (4.4–4.7 eV). In this material, however, metal ions penetrate into the photo-active layer after

the indium tin oxide (ITO) is corroded by the strongly acidic PSS in the PEDOT:PSS. As a result, due to these metal ions in the photo-active layer acting as quenching sites for dissociated carriers, the long-term stability is reduced [18–20]. In response to this, there have been studies to improve the PCE and stability without using PEDOT:PSS as an anode buffer layer.

Helander et al. reduced the work function of ITO to 6.1 eV by Cl doping, and achieved power efficiency more than twice as high as that of conventional organic light-emitting diodes (OLEDs), by fabricating chlorinated ITO (Cl-ITO)-based OLEDs [21]. However, according to Sun et al., the photovoltaic performance of PSCs with Cl-ITO degrades quickly with time. This is attributed to the reduction of the work function caused by the desorption of Cl from the ITO surface [22]. To fabricate more stable PSCs, there have been studies on the formation of a monolayer using molecules that could be covalently bonded to the substrate surface [23,24].

Kim et al. controlled the work functions of ITO by introducing ITO to a self-assembly monolayer (SAM), which has an electron-donating or withdrawing group as a terminal group [25]. Trichloro(3,3,3-trifluoropropyl)silane was used as a precursor of CF₃-SAM to enable covalent bonding to the substrate surface. A hydroxyl group was formed through Ar-plasma treatment of the ITO surface, and the ITO of the CF₃-ITO fabricated had an increased work function of with an electron withdrawing group. The PCE of the PSC was greater than that of the device without PEDOT:PSS, but there has been no comparison between a CF₃-ITO-based device and a PEDOT:PSS-based device. In addition, the stability characteristics have not been reported either.

In general, as the number of fluorines in the SAM-treated ITO increases, so does the work function [26,27]. To match the work

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function of ITO with the highest occupied molecular orbit (HOMO) level (6.2 eV) of the acceptor, phenyl-C₆₁-butyric acid methyl ester (PCBM), triethoxy(1H,1H,2H,2H-perfluoro-1-octyl)silane (FOTES) is used as a SAM precursor. FOTES has abundant fluorine, and rarely experiences desorption from the ITO surface, since it is covalently bonded. In addition, if a catalyst such as nitric acid is used for SAM treatment, it is possible to form a homogeneous monolayer. Therefore, FOTES-ITO has been fabricated and characterized, and the device performance and stability were measured by fabricating FOTES-ITO-based PSCs.

2. Materials and measurements

2.1. Materials

ITO glass from Samsung Corning was used as a transparent electrode (ITO: 170 nm, 10 Ω /sq). PEDOT:PSS (AI 4083) was purchased from Clevis, and poly(3-hexylthiophene) (P3HT), which was used as a donor material in the photoactive layer, was purchased from Rieke metal. PCBM, the acceptor material, was purchased from Nano C. FOTES was bought from Aldrich and used as a SAM precursor. Nitric acid (HNO₃, 61%) was obtained from Dae Jung Chemical & Metals and used as a catalyst. The chemical structure of P3HT, PCBM, and FOTES are shown in Fig. 1(a–c).

2.2. Measurements

All of the thin films were fabricated using a GMC2 spin coater (Gensys), and their thicknesses were measured using an alpha step 500 surface profiler (KLA-Tencor). The morphology of the FOTES-ITO was observed through atomic force microscopy (AFM, PSIA XE-100). The contact angle was measured with a contact-angle meter (KRUSS K6). The work function of ITO and FOTES-ITO were measured and calculated by ultraviolet photoelectron spectroscopy (UPS). UPS spectra were obtained using monochromatized He I radiation ($h\nu=21.22$ eV) from a He-resonance lamp, with an energy resolution of 0.03 eV, as determined from the slope of

the Fermi edge of a sputter-cleaned gold sample. The current density–voltage (J - V) characteristics of the PSCs were measured using a Keithley 2400 source measure unit. The devices were evaluated at 298 K using a Class A Oriel solar simulator (Oriel 96000 150-W solar simulator) with a xenon lamp that simulates AM 1.5 G irradiation (100 mW/cm^2) from 400 to 1100 nm. The instrument was calibrated with a monocrystalline Si diode fitted with a KG5 filter to bring the spectral mismatch to unity. The calibration standard was calibrated by the National Renewable Energy Laboratory (NREL). The incident photon-to-current efficiency (IPCE) (Mc science) was measured against the best-performance device. All devices were encapsulated and stored in air for 600 h in the dark for investigation of the long-term stability (Test ID: ISOS-D-1Shelf) [28].

3. Experimental

3.1. Cleaning of patterned ITO glass

To clean the patterned ITO glass, it was sonicated for 20 min, sequentially in detergent (Alconox[®] in deionized water, 10%), acetone, isopropyl alcohol, and deionized water. The moisture was thoroughly removed by N₂ gas flow. To ensure complete removal of all of the remaining water, the patterned ITO glass was heated on a hot plate for 10 min at 100 °C. For hydrophilic treatment of the patterned ITO glass, it was cleaned for 10 min in a UVO cleaner.

3.2. SAM formation on ITO

FOTES (5 mM) and nitric acid (0.01 M) were mixed at a 100:1 M ratio. The mixed solution was used after being filtered with a 0.2- μm PTFE syringe filter. To form an OH group on the surface of the patterned ITO glass, UVO cleaning was performed for 30 min. After applying 0.4 ml of FOTES solution to the patterned ITO glass, it was kept on a petri dish at 25 °C for 17 h. Then, it was sonicated with isopropyl alcohol for 3 min and annealed at 90 °C on a hot plate in an N₂-filled glove box.

3.3. Fabrication of PSCs

To produce a conventional cell, an aqueous solution of PEDOT:PSS was spin-coated to form a 40-nm-thick film on the patterned ITO glass. The substrate was dried for 10 min at 140 °C in air and then transferred into a glovebox to spin coat the active layer. A solution containing a mixture of P3HT/PCBM (1:0.6) in ODCB was then spin-coated on top of the PEDOT:PSS to produce a 130-nm-thick photoactive layer, which was then subjected to thermal annealing at 160 °C for 10 min. To form the metal cathode, Al (5 $\text{\AA}/\text{s}$, 100 nm) was thermally deposited in a high-vacuum ($< 10^{-7}$ Torr) chamber.

For the FOTES-ITO-based PSC, photo-active ink was then spin-coated on top of the FOTES-ITO to produce a 130-nm-thick photoactive layer, which was then thermally annealed at 160 °C for 10 min. The method of forming the metal electrode was the same as that for the conventional cell.

4. Results and discussion

Fig. 2 shows the J - V curve and IPCE data of the PSCs fabricated with FOTES-ITO, ITO/PEDOT:PSS, and ITO with no surface treatment. The results are presented in Table 1. The PCE of the FOTES-ITO-based PSC was 3.4%, showing better efficiency than the device in which PEDOT:PSS was used as an anode buffer layer. This result was

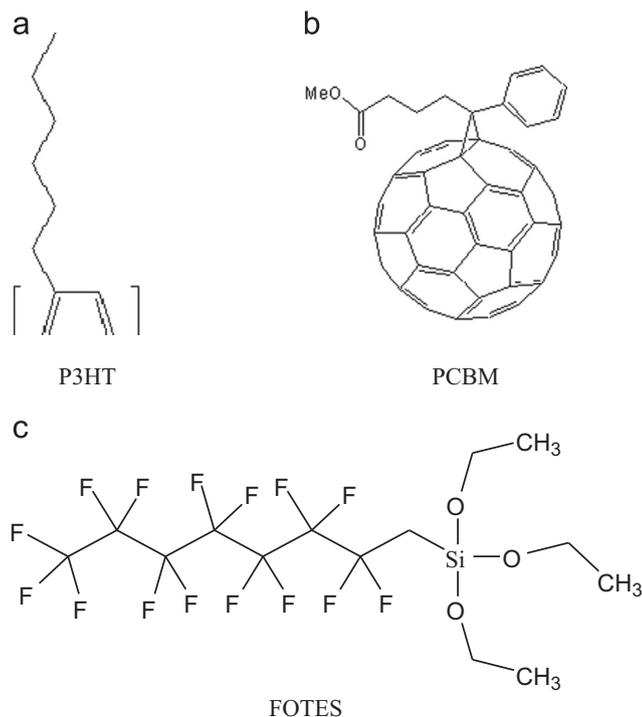


Fig. 1. Chemical structure of (a) P3HT, (b) PCBM and (c) FOTES.

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