

Effective protection of sequential solution-processed polymer/fullerene bilayer solar cell against charge recombination and degradation



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ABSTRACT

Both charge recombination and degradation in sequential solution processed polymer/fullerene bilayer organic photovoltaics (OPV) are effectively reduced by the insertion of a TiO₂ inter-layer between the bilayer and Al electrode. The polymer/fullerene bilayer composed of a poly(3-hexylthiophene) (P3HT) bottom-layer and a [6,6] phenyl C61-butyric acid methyl ester (PCBM) top-layer shows significant change in morphology due to the substantial inter-penetration of P3HT and PCBM during the thermal annealing process. Consequently, the bilayer surface becomes P3HT rich resulting in significant charge recombination at the bilayer/Al interface of the bilayer OPV. The charge recombination rate of the bilayer OPV is reduced by one order of magnitude upon the insertion of a TiO₂ nanoparticle inter-layer between the bilayer and the Al electrode after the thermal annealing process. In contrast, when the thermal annealing process is conducted after insertion of the inter-layer, the effect of the TiO₂ inter-layer becomes insignificant. The V_{OC} and efficiency of the bilayer OPV is greatly enhanced from 0.37 to 0.66 V and 1.2% to 3.7%, respectively by utilizing the properly constructed TiO₂ inter-layer in the bilayer OPV. Additionally, insertion of the TiO₂ inter-layer significantly improves the stability of the bilayer OPV. The bilayer OPV with a TiO₂ inter-layer maintains 51% of its initial PCE after storage under dark ambient conditions for 700 h without encapsulation, whereas the bilayer OPV without a TiO₂ inter-layer did not show any solar cell performance after 200 h under the same conditions.

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1. Introduction

Organic photovoltaic (OPV) devices mainly utilize the bulk heterojunction (BHJ) type photo-active layer. A polymer:fullerene blend solution deposition (BSD) process is normally used to prepare the BHJ type active layer. The BSD process is effective in forming a heterojunction with a large surface area in a single photo-active layer. The sequential solution deposition (SqD) process, wherein an electron donating layer is deposited first followed by an electron accepting layer, can be utilized to form the heterojunction in bilayer OPV devices. The SqD process is more straightforward facilitating facile device optimization compared to the one-step blended solution process. Recently, several interesting reports on SqD processed bilayer OPVs based on P3HT and fullerene derivatives have been reported [1–5].

Thermal annealing has proven to be one of the most effective ways to enhance the bilayer OPV efficiency by increasing the heterojunction area and thus improving the exciton dissociation

between the donor and acceptor [1,6]. Schwartz et al. claimed that the bilayer OPV provides a reduced carrier recombination and a higher degree of control [2]. Burn et al. reported that the SqD processed P3HT/PCBM bilayer produced an intermixed heterojunction even without thermal annealing [4]. PCBM is able to thoroughly diffuse into the P3HT layer upon thermal annealing, and the bilayer OPV showed superior solar cell performance compared to the BHJ OPV prepared by the BSD process. Heeger et al. reported that the bilayer OPV showed nearly identical nano morphologies with the BHJ OPV due to the penetration of PCBM into the P3HT layer [7]. However, Schwartz et al. claimed that thermal annealing improved the performance of the bilayer OPV not because of PCBM penetration into the P3HT but because annealing improved the PCBM crystallinity [8]. There are controversial opinions regarding PCBM penetration during thermal annealing. In addition, further studies regarding the stability of the SqD processed bilayer OPV are needed.

In this study, the change in the surface morphology of the P3HT/PCBM bilayer induced by the thermal annealing process was investigated. It was found that the thermal annealing process caused a significant change in the morphology of the bilayer that unfavorably changed the surface composition of the bilayer leading

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to a significant charge recombination at the bilayer/Al electrode interface. A solution processed TiO₂ nanoparticle inter-layer was utilized to reduce the charge recombination, and the effects of the TiO₂ inter-layer on the solar cell performances of the bilayer were investigated for the different thermal annealing processes (pre- or post-annealing). Furthermore, the effect of the TiO₂ inter-layer on the stability of the bilayer OPV was investigated.

2. Experimental

2.1. Synthesis and characterization of TiO₂ nanoparticles

To a solution of 10.27 mL acetylacetone in 38 mL *n*-butanol that had been stirring for 15 min at room temperature was slowly added 34 mL of titanium butoxide with stirring. After the exothermic reaction was complete, the solution was cooled to room temperature and a solution of 3.80 g of *p*-toluene sulfonic acid dissolved in 17.65 mL deionized water was slowly added with continuous stirring. After an hour, the reaction mixture was stirred at 60 °C overnight. The resulting solution was precipitated with 100 mL toluene and centrifuged at 2000 rpm. The TiO₂ nanoparticle powder thus attained was dried in a vacuum chamber and stored under ambient air.

Transmittance electron microscopy (TEM) was used to characterize the synthesized TiO₂ nanoparticles. The TEM image was obtained using a Tecnai™ G2 F20 (FEI, USA) with a 200 kV accelerating voltage. The TEM image of the TiO₂ NPs is shown in Fig. 1(a), and the average nanoparticle size was 3–5 nm. The structure of the synthesized TiO₂ particles was investigated by X-ray diffraction (XRD). The XRD patterns for samples deposited on a glass slide were taken with a Philips PW 1830 diffractometer with a Ni-filter Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$) operated at 40 kV and 20 mA. Strong diffraction directions at (101) and (200) (Fig. 1(b)) indicated that the TiO₂ particles were in the anatase form.

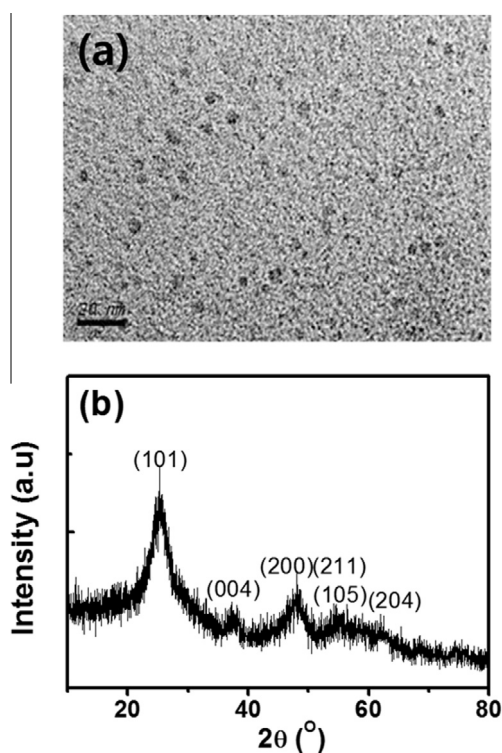


Fig. 1. (a) TEM image and (b) XRD pattern of the TiO₂ nanoparticles.

2.2. Device fabrication

The bilayer OPVs were fabricated with the structure Glass/ITO/PEDOT:PSS/bilayer/Al or Glass/ITO/PEDOT:PSS/bilayer/TiO₂/Al. To fabricate the bilayer devices, tin doped indium oxide (ITO) coated glass substrates were first prepared by cleaning in an ultrasonic bath of isopropyl alcohol, followed by acetone, and finally isopropyl alcohol again for 10 min each. After completely drying the substrates, they were then exposed to UV-ozone for 20 min to prepare the substrates for spin coating with poly(3,4-ethylene dioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) (Baytron P VP Al 4083). PEDOT:PSS was spin-coated at a speed of 4000 rpm and then dried in a vacuum oven at 110 °C for 10 min. P3HT dissolved in chlorobenzene at a concentration of 15 mg/mL was then deposited by spin-coating at 1000 rpm followed by drying in the air for 10 min. Subsequently, PCBM dissolved in dichloromethane (4 mg/mL) was spin-coated on top of the P3HT layer at 4000 rpm. The bilayer film was pre-annealed at 150 °C for 20 min in a glove box ([BL]_A). TiO₂ nanoparticles dispersed in butanol (0.4 wt%) were deposited by spin-coating at 1500 rpm onto the [BL]_A film to construct the [BL]_A/TiO₂ film. The [BL]_A/TiO₂ film was dried at 60 °C for 30 min in a N₂ filled glove box. For comparison with a pre-annealed bilayer film, two different types of post-annealed bilayer structures were fabricated. One bilayer was thermally annealed after construction of the BL/TiO₂ film ([BL/TiO₂]_A), and another was thermally annealed after constructing the bilayer, the TiO₂ inter-layer, and the Al electrode layer ([BL/TiO₂/Al]_A). The Al electrode was deposited under vacuum at a pressure of $\sim 10^{-6}$ Torr at a thickness of ~ 100 nm.

To investigate the extent of the PCBM inter-penetration into the P3HT layer, a bilayer OPV with an inverted structure of ITO/TiO₂/P3HT/PCBM/PEDOT:PSS/Ag was fabricated. A 0.4 wt% TiO₂ nanoparticle solution in butanol was spin-coated on a cleaned ITO substrate at 3000 rpm and dried in the air for 1 h at 60 °C. Construction of the BL was completed using the SqD process. For the BHJ layer, a blend solution of P3HT:PCBM (1:0.6 wt%) with a concentration of 24 mg/mL in chlorobenzene was spin-coated at 1000 rpm for 15 s. A PEDOT:PSS: isopropyl alcohol (1:1) solution was spin-coated onto the active layer at a speed of 5000 rpm for 1 min. The BL and BHJ active layers were thermally annealed at 150 °C for 20 min in a N₂ filled glove box prior to transfer to a vacuum chamber to deposit the Ag electrode. A 100 nm Ag electrode was deposited on top of the PEDOT:PSS layer by thermal evaporation ($\sim 10^{-6}$ Torr).

2.3. Measurement

Current density–voltage (*J*–*V*) characteristics were measured under AM 1.5G irradiation (100 mW cm⁻²) or in the dark with a Keithley 2400 Source Measure Unit. A class-A solar simulator (McScience, Korea) equipped with a 150 W Xenon lamp was used as a light source, and anNREL-calibrated mono Si solar cell with a KG-3 filter was used to adjust the AM 1.5G one sun light intensity.

The water contact angle measurement was conducted using a KSV CAM 101. The transient photovoltage (TPV) measurements were performed at steady state under continuous illumination from an intensity adjustable white LED. When the device output reached a steady state, the device was perturbed using a green LED with a 1 Hz pulse rate. The resulting voltage transient was acquired by a TDS3054B Tektronix digital oscilloscope with a 1 M Ω input impedance. TPV results were fitted to a mono exponential decay function in order to find the carrier recombination lifetime [9].

The PL spectra of the films were obtained using a Perkin Elmer LS 55. Atomic force microscopy (AFM) images were obtained in non-contact mode using an XE-70 (Park Systems Corp., Korea).

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