



Wide range thickness effect of hole-collecting buffer layers for polymer:fullerene solar cells



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ABSTRACT

Here we report the wide range thickness effect of hole-collecting buffer layers (poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)) on the performance of polymer:fullerene solar cells with blend films of poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PC₆₁BM). The thickness of the PEDOT:PSS layers was controlled from 3 nm to 625 nm, followed by characterizations such as optical transmittance, electrical resistances in the in-plane and out-of-plane directions, work functions, contact angles, and device performances. Results showed that the optical transmittance was gradually decreased with the PEDOT:PSS thickness but a maximum value was measured for other properties in the thickness range of 10–30 nm. The device performance was noticeably improved with only 3 nm-thick PEDOT:PSS layer, while it was almost similar in the thickness range of 30–225 nm in the presence of gradual decrease in the surface roughness. The similar device performance between 30 nm and 225 nm has been assigned to the compensation effect between the reduced electrical resistance (increased conductivity) and the decreased optical transmittance as the thickness of the PEDOT:PSS layer increased.

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1. Introduction

Polymer solar cells have recently attracted keen attention because of their potential for mass production of low-cost plastic solar modules that can be used for both outdoor and indoor energy harvesting medium in a type of various shapes [1–4]. The viability of low-cost production of polymer solar cells can be attributed to the room-temperature coating processes of polymeric layers that are sandwiched between electrodes because polymer solu-

tions are used in the coating step [5–7]. In addition, continuous roll-to-roll process using a plastic film substrate can be another big benefit to achieve the low-cost manufacturing of polymer solar cells [8,9].

Interestingly, to date, polymer solar cells that exhibit relatively higher power conversion efficiencies (PCE > 4%) have been made by employing bulk heterojunction (BHJ) films of electron-donating polymers and soluble fullerenes [10–16]. Three organic materials such as electron-donating (main light absorber) polymers, electron-accepting fullerenes, and hole-collecting polymers can be classified as a core part for the BHJ type polymer:fullerene solar cells [17–20]. As a hole-collecting polymer, poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) has been widely used because of its work function (~5.2 eV) that can help reduce the energy barrier for hole carrier

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transport between BHJ active layers and electrodes [21–25].

Considering the limited conductivity of the PEDOT:PSS material (up to ~ 500 S/cm), the thickness-dependent conductivity is expected for the PEDOT:PSS layer. This implies that the performance of polymer solar cells can be affected by varying the thickness of the PEDOT:PSS layer. On this account, a couple of studies have been reported for the thickness effect of the PEDOT:PSS layer on the performance of polymer solar cells [22,26–29]. However, the thickness range studied by these previous studies was relatively narrow, which was insufficient to understand the wide range thickness effect of the PEDOT:PSS layer.

In this work, we tried to investigate the wide range thickness effect of the PEDOT:PSS layer on the performance of polymer:fullerene solar cells by varying the thickness of the PEDOT:PSS layer from 3 nm to 625 nm and attempted to correlate the device performances with various characteristics of the PEDOT:PSS layers such as optical transmittance, in-plane (IP) and out-of-plane (OOP) electrical resistance, work function, and contact angle. As a BHJ layer, the blend film of poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl- C_{61} -butyric acid methyl ester ($PC_{61}BM$) has been employed because the P3HT: $PC_{61}BM$ film has been typically used as one of the standard organic solar cells so far.

2. Materials and methods

PEDOT:PSS solutions (conductivity = ~ 500 S/cm, PH500) were purchased from HC Starck and are subject to dilution with isopropyl alcohol for thickness control. P3HT (regioregularity = 92%; weight-average molecular weight = 72 kDa) and $PC_{61}BM$ (purity = 99.5%) were used as received from Rieke Metals and Nano-C, respectively. Blend solutions of P3HT and $PC_{61}BM$ (P3HT: $PC_{61}BM$ = 1:0.7 by weight) were prepared using chlorobenzene (solvent) at a solid concentration of 51 mg/ml. The solutions were vigorously stirred on a hot-plate stirrer for well-mixing just before spin-coating process.

Prior to the film and device fabrication, indium-tin oxide (ITO)-coated glass substrates were patterned with a photolithography/etching technique. The patterned ITO-glass substrates were cleaned by a wet-process using acetone and isopropyl alcohol, followed by UV-ozone treatment. On top of the cleaned ITO-glass substrates the PEDOT:PSS layers were spin-coated by varying solution concentrations at 2500 rpm. The coated PEDOT:PSS samples were subject to thermal annealing at 230 °C for 15 min. The thickness of the coated PEDOT:PSS layers was 3, 7, 10, 20, 30, 40, 50, 85, 130, 225, and 625 nm. Next, the BHJ (P3HT: $PC_{61}BM$) layer was spin-coated on the PEDOT:PSS layer-coated ITO-glass substrates at 1500 rpm. The BHJ layer-coated samples were soft-baked at 60 °C for 15 min and their thickness was ca. 170 nm. These samples were moved into a vacuum chamber system equipped inside a nitrogen-filled glove box for the deposition of top electrodes (aluminum – Al). The thickness of the Al electrodes was controlled to be 100 nm, while the active area of devices was 0.09 cm² (device structure: glass/ITO/PEDOT:PSS/P3HT: $PC_{61}BM$ /Al). Finally the devices were ther-

mally annealed at 140 °C for 30 min inside the nitrogen-filled glove box and stored in the same glove box before measurement. For the measurement of OOP (normal to the film thickness) direction resistance, the Al electrodes were directly deposited on top of the PEDOT:PSS layers that were coated on the ITO-glass substrates (device structure: glass/ITO/PEDOT:PSS/Al).

The optical transmittance of the PEDOT:PSS-coated ITO-glass substrates was measured using an UV-visible spectrometer (Optizen 2120UV, MECASYS). The IP (parallel to the film plane) direction resistance of the PEDOT:PSS layer was measured using a four-probe measurement system equipped with an semiconductor parameter analyzer (4200-SCS, Keithley), while the OOP direction resistance was obtained from the current–voltage (I – V) curves of the glass/ITO/PEDOT:PSS/Al devices which were measured using an electrometer (2400, Keithley). The work function of the PEDOT:PSS layers that are coated on the ITO-glass substrates or bare glass substrates was measured using a photoelectron yield spectrometer (AC-2, Riken-Keiki). The solar cell performance of devices was measured using a specialized solar cell measurement system equipped with a solar simulator (Air Mass 1.5G, 100 mW/cm², Newport Oriol) and an electrometer (2400, Keithley). The surface morphology of the ITO and PEDOT:PSS layers was measured using an atomic force microscope (AFM, Nano Xpert-II, EM4SYS).

3. Results and discussion

The device structure with the PEDOT:PSS layer is illustrated in Fig. 1a. Because solar light should pass through the PEDOT:PSS layer-coated ITO-glass substrates toward the light-absorbing layer (P3HT: $PC_{61}BM$), the optical transmittance of the PEDOT:PSS layer is expected to sensitively affect the photocurrent generation. As shown in Fig. 1b, the optical transparency of the PEDOT:PSS layer-coated ITO-glass substrates was changed with the thickness of the PEDOT:PSS layer. A noticeable change in the transparency can be found from around 85 nm (thickness) with naked eyes, while the characters “KNU” on the background sheet were considerably blocked at the thickness of 625 nm. To further investigate the optical transmittance trend according to the wavelength of incident light, we measured the optical absorption spectra of the PEDOT:PSS layer-coated ITO-glass substrates and converted these absorption spectra to the transmittance spectra.

As shown in Fig. 2a, the pristine ITO-glass substrates without any PEDOT:PSS layer showed a minimum transmittance at around 430 nm (wavelength) and a maximum transmittance at around 600 nm (wavelength), which can be attributed to the different refractive index and Fresnel reflection effects depending on the wavelength of incident light [30]. Here we note that the extremely low transmittance below 350 nm (wavelength) for the pristine ITO-glass substrate is ascribed to the strong absorption of glass substrates. As the thickness of the PEDOT:PSS layer increased, the minimum and maximum transmittances were changed owing to the varied optical attenuation condition [31]. In particular, a huge change in the spectral shape was

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