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Semitransparent Organic Solar Cells with Polyethylenimine Ethoxylated Interfacial Layer Using Lamination Process

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SUMMARY Semitransparent organic solar cells were fabricated using lamination process. The devices were realized by using two independent substrates with transparent indium-tin-oxide electrode. One substrate was coated with poly(ethylenedioxy-thiophene)/poly(styrenesulfonate) layer and active layer of poly(3-hexylthiophene-2,5-diyl) (P3HT) and (6,6)-phenyl-C61 butyric acid methyl ester mixture. Another substrate was coated with ultra-thin polyethylenimine ethoxylated. The two substrates were laminated using hot press system. The device exhibited semitransparency and showed typical photovoltaic characteristics with open voltage of 0.59 V and short circuit current of 2.9 mA/cm².

key words: organic solar cell, bulk hetero structure, interfacial layer, lamination process, semitransparent

1. Introduction

Organic solar cells (OSCs) are actively investigated as sustainable energy source for next generation [1]–[4]. The fabrication of OSCs often utilizes the solution process. Therefore, OSCs are expected to have advantages over solar cells such as Si in terms of energy payback time and cost payback time. The efficiency is advanced daily by introducing bulk heterojunction structures [2]–[4] with p-type and n-type materials and organic-inorganic hybrid perovskite materials [5]. However, the adoption of OSCs requires further efficiency and cost reduction. In order to improve the efficiency of solar cells, improvement of open voltage, short circuit current and fill-factor is required. About the open voltage, it is necessary to increase the difference between the highest occupied molecular orbital level of the donor and lowest unoccupied molecular orbital level of the acceptor and increase the difference between the anode and cathode work functions. Transparent oxide electrodes such as indium-tin-oxide and fluorine doped tin oxide are used for OSCs because at least one of the anode and cathode is transparent. Semitransparent OSCs can be realized to use transparent electrodes both anode and cathode. However, since the use of the same transparent electrode does not have a work function difference, a high open voltage cannot be expected. Recently, a decrease in the work function has been reported by modifying using a polyethyleneimine and the derivative on the ITO surface [6], [7]. On the other hand, lamination method is expected as a process technology for cost reduction [8]–[13]. In this paper, we have re-

ported semitransparent OSCs with transparent anode and transparent cathode modified polyethylenimine ethoxylated interfacial layer using lamination process.

2. Experimental

Figure 1 shows the device structure and organic materials under study. Indium-tin-oxide (ITO)-coated glass substrates were treated by scrubbing, ultrasonic cleaning, and UV-ozone treatment before film coating. Poly(ethylenedioxythiophene)/poly(styrenesulfonate) (PEDOT:PSS) was spin-coated on an ITO substrate for anode and was baked at 200°C for 5 min in air. Poly(3-hexylthiophene-2,5-diyl) (P3HT) and (6,6)-phenyl-C61 butyric acid methyl ester (PCBM) blend as an active layer were coated using spin-coating on PEDOT:PSS layer. The ratio of P3HT:PCBM was 1:1 by weight and the blend dissolved in chloroform. After spin-coat, the substrate was baked at 200°C for 5 min in N₂ environment. On the other hand, polyethylenimine ethoxylated (PEIE) water-solution diluted by ethanol was spin-coated on another ITO substrate for cathode. Part of PEIE layer was spin-rinsed with ethanol for 2 times. The both substrates were laminated using a heater press system (Mikado Technos, MKP-150PV-WH). The pressure of lamination was 0.5 MPa under 200°C condition. The device structure was glass substrate/ITO/PEDOT:PSS/P3HT:PCBM/PEIE/ITO/glass substrate. Active area was 2 × 2 mm². The voltage-current characteristics were measured using a semiconductor pa-

Fig. 1 Device structure and molecular structure used in this study.

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rometer analyzer (HP4155B). The light source for measurement was a solar simulator (Yamashita Denso, YSS-E40) at a light intensity of AM1.5, 100 mW/cm². The absorption and transmission spectra were measured by spectrophotometer (Hitachi, U-1900). Sensitivity spectra were measured using a motor controlled monochromator (Shimadzu, SPG-100ST, AT-100PCC, and AT-100PL), a halogen lamp (Scott, MegaLight 100), and a digital electrometer (Advantest, TR8652). Sensitivity was calibrated using a Si photodiode (Hamamatsu photonics, S1223).

3. Results and Discussion

Laminated devices were adhered without detachment during handling. This suggests that the polymer layer functions as an adhesive layer by heating and pressurization. Figure 2 show the device photograph of the laminate device and the transmission spectrum for the device area. It was shown to be red because of light absorption by the P3HT:PCBM layer, however the characters laid under the device was clearly confirmed and transparency was confirmed. Transmittance of over 50% was obtained above 640 nm wavelength. Lower transmittance below 640 nm wavelength was originated absorption of P3HT:PCBM layer.

Figure 3 show the J - V characteristics obtained under illumination of AM1.5. Device characteristics were dramatically improved to coat ultra-thin PEIE layer on ITO for cathode. The open voltage (V_{oc}) increased from 0.31 V to 0.59 V by inserting PEIE. It has been reported that the work function of ITO surface decreases by coating the PEIE

Fig. 2 Device photograph of the laminate device and the transmission spectrum for the device area.

layer [12], [13]. The improvement of the open voltage is due to the increase in the work function difference between the anode and cathode. The conversion efficiency was 6 times and short circuit current (J_{sc}) was more than twice as high as that of J_{sc} .

Figure 4 shows the comparison with a device fabricated by vacuum deposition cathode of LiF/Al stack. The J_{sc} , the V_{oc} , the fill factor (FF), and efficiency (η) of each device were 0.59 V, 2.9 mA/cm², 0.38, 0.65% for lamination devices and 0.62 V, 3.7 mA/cm², 0.43, 1.01% for evaporation device, respectively. Almost same value was obtained for V_{oc} , however, J_{sc} was decreased in the laminate device. The cause is the imperfection of the laminate interface formation state and the decrease in the bulk hetero interface. After laminating, some uneven interference fringes are seen, and optimization of spin coating and laminate conditions is required. The ratio of photo-conductivity to dark-conductivity σ_R for lamination and evaporation devices was 1.2×10^5 and 2.3×10^4 at the voltage of -1 V, respectively. This high σ_R suggests no damage to the organic

Fig. 3 J - V characteristics of the devices with and without poly-ethyleneimine interfacial layer under illumination.

Fig. 4 J - V characteristics of the lamination devices compared with evaporation device under illumination.

Fig. 5 (a) Absorption spectrum of the device area and (b) sensitivity spectrum.

layer/electrode interface due to laminates. The characteristics of the devices stored in air for 100 days were also evaluated. Unsealed evaporation devices had no device operation after 100 days, while lamination devices were showed the initial 88% characteristics. This means that the glass plates on both side of laminated device blocked water and oxygen penetration, and enhanced the device life.

Figure 5 shows absorption spectrum for the device area and photo sensitivity versus wavelength properties. Absorption is observed at wavelength less than 650 nm due to P3HT and PCBM and peaks at 500 nm. It is also seen that power generation occurs in the same wavelength range as the absorption of the device.

4. Conclusion

In summary, we fabricated semitransparent organic solar cells using lamination process. Laminated devices were adhered without detachment during handling. The device exhibited semitransparency over 50% above 640 nm wavelength. Device characteristics were dramatically improved to coat ultra-thin PEIE layer on ITO for cathode and were showed typical photovoltaic characteristics with open voltage of 0.59 V and short circuit current of 2.9 mA/cm².

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