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Fully spray-coated ITO-free organic solar cells for low-cost power generation

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ABSTRACT

We report on cost-effective ITO-free organic solar cells (OSCs) fabricated by a spray deposition method. All solution-processable layers of solar cells—a highly conductive poly(3,4-ethylenedioxythiophene):-poly(styrenesulfonate) (PEDOT:PSS) layer and a photoactive layer based on poly(3-hexylthiophene) (P3HT) and 1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C_{61} (PCBM)—were spray-coated. PEDOT:PSS anode films with various thicknesses were prepared by controlling the spray deposition time. The transmittance and sheet resistance of PEDOT:PSS anodes were varied from 89.0% to 67.4% and from 358 to 63.3 Ω/squares, respectively, corresponding to an increase in film thickness. The best device exhibited a high power conversion efficiency of 2.17% under 100 mW cm^{-2} illumination with air mass (AM) 1. 5 global (G) condition. More importantly, the efficiency of the fully spray-coated OSC with the PEDOT:PSS anode was comparable to that of conventional ITO-based devices, demonstrating the feasibility of fabricating all-spray-deposited OSCs without a conventional spin-coating method and the possibility of replacing the costly vacuum-deposited indium tin oxide (ITO) with highly conductive polymer films fabricated by inexpensive spray deposition techniques.

1. Introduction

Conjugated polymer-based organic solar cells (OSCs) have received significant attention due to their potential for low-cost roll-to-roll manufacturing and large-area processability on flexible substrates [1–11]. Among the available polymer solar cell systems, poly(3-hexylthiophene) (P3HT) and 1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C_{61} (PCBM) blends show efficiencies of up to 4–5% [8,9]. In addition, with the aid of novel donor polymer, the power conversion efficiencies of bulk-heterojunction (BHJ) solar cells have continuously increased up to ~6% [12,13]. Furthermore, various approaches to realize an efficient OSC with roll-to-roll processing, large-area processability, and excellent stability on low-cost and flexible substrates have been actively investigated [14–17]. As a result, the continual development in efficiency, cost, process, and stability makes polymer-based organic solar cells more attractive as a cost-effective solution to today's energy-shortage problems [1–3].

The realization of low-cost, flexible, and highly efficient OSCs based on cost-effective roll-to-roll (RTR) processing technologies is an obvious goal in this field [18–20]. For this reason, several research groups have explored various printing or coating techniques such as screen printing [21], doctor blading [22], brush painting [23], and inkjet printing [24] to form the photoactive layer of OSCs. For the same purpose, we recently introduced spray coating to efficiently deposit the active layer from solution and demonstrated that the efficiency of OSCs with spray-coated active layers was comparable with that of devices with spin-coated active films [25]. Due to the fast processing and compatibility with roll-to-roll manufacturing, many research groups have investigated the spray deposition as a cost-effective coating method for photoactive layers of OSCs [26–30]. In addition, the spray process was expended to deposit poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) layers [29–31]. In particular, most recently, fully spray-coated organic photodiodes and solar cells that include the spray-coated hole-transporting PEDOT:PSS layer and the spray-coated P3HT:PCBM layer were reported and demonstrated that the spray process is a practicable coating technology to deposit all solution used in organic devices [29,30]. The organic-based hole-transporting and photoactive layers that were created using a simple and fast solution-based process may be highly desirable for the realization of low-cost and high-efficiency OSCs. However, until very recently, only a few approaches for fully spray-coated OSCs

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have been reported, and fully spray-coated OSCs reported so far have been generally fabricated on expensive crystalline indium tin oxide (ITO) electrode prepared at cost-intensive vacuum processes and high temperatures. Considering the high cost of indium and limited flexibility of ITO, the ITO electrode cannot be considered an ideal electrode material for low-cost, flexible, and high-efficiency OSCs. For this reason, various modified forms using PEDOT:PSS and metal grids and their introduction to ITO-free devices are all being actively investigated [32–36].

In this study, we demonstrate on cost-effective ITO-free organic solar cells fabricated by a spray deposition method. All solution-processable layers of solar cells—the highly conductive PEDOT:PSS anode layer and the photoactive P3HT:PCBM layer—were spray-coated. Here, we investigated a recently developed PEDOT:PSS formulation, Baytron PH 500 provided by H.C. Starck, as a polymer anode for fully spray-coated OSCs. This formulation recently demonstrated a capacity for substitution of the ITO electrode by PH 500 films modified by the addition of polar solvents to obtain a conductivity as high as ~500 S/cm [37,38]. Fully spray-coated OSCs with PEDOT:PSS anode films present a high power conversion efficiency that is comparable to that of ITO-based reference cells, demonstrating the feasibility of fabricating all-spray-deposited OSC without a conventional spin-coating method and the possibility of replacing the costly vacuum-deposited ITO with highly conductive polymer films fabricated by inexpensive spray deposition techniques.

2. Experimental

Fig. 1 shows the fabrication sequence for ITO-free bulk-heterojunction solar cells with a spray-coated PEDOT:PSS anode and a spray-coated P3HT:PCBM active layer fabricated using a simple spray deposition; the chemical structures of materials used for the fabrication of ITO-free cells are also shown. To fabricate OSCs without and with ITO, glass (Superior Marienfeld, Germany) and ITO-coated glass (Samsung Corning Co, Ltd., ~10 Ω/square) substrates were cleaned with a detergent (Mucasol, Merz) followed by ultrasonication in acetone and isopropyl alcohol. Each substrate was then treated with UV/ozone to improve the wettability of PEDOT:PSS. The highly conductive polymer anode first layer was prepared by addition of 5% dimethylsulfoxide (DMSO) to an aqueous solution of PH 500 [37]. The modified PH 500 solution was transferred to a handheld airbrush (Fenghua Bida, BD-130) and sprayed at a pressure of 0.1 MPa and at a distance of 20 cm, nozzle-to-substrate, in air, followed by drying at 120 °C for 20 min in air, as shown in Fig. 1. PEDOT:PSS anode films with various thicknesses were prepared by controlling the spray deposition time. The second layer, a bulk-heterojunction film composed of interconnected networks of electron-donor and -acceptor materials, was prepared by spray-coating a mixture of P3HT (Rieke Metal) and PCBM (Nano C). For the fabrication of the photoactive layer, a solution of 30 mg of P3HT and 15 mg of PCBM in 20 ml of chlorobenzene was sprayed at a pressure of 0.1 MPa and at a distance of 20 cm, nozzle-to-substrate, for 20 s in air, which formed the active layer with a thickness of ~110 nm [25]. After the active layers were spray-coated on top of the PEDOT:PSS layer, thermal annealing was carried out at 110 °C for 10 min on a hot plate in N_2. Finally, the reflective metal cathode consisting of 100 nm Al on top of 20 nm Ca was thermally evaporated through a shadow mask to produce an active area of 4.66 mm^2 in a vacuum with a pressure of 10^{-6} Torr. For comparison, two reference ITO-based solar cells using spin-coating a standard VPAI 4083 and the modified PH 500 solutions to be used as an interfacial layer in ITO-based cells were also fabricated using experimental procedures identical to those used for fabrication of fully spray-coated ITO-free cells. VPAI 4083 and modified PH 500 solutions were spin-coated at 5000 rpm for 40 s and were subsequently annealed at 120 °C for 20 min. The UV–vis transmission measurements were performed using a Perkin–Elmer Lambda 12 UV/vis spectrophotometer. The sheet resistance was measured using a standard four-point-probe system with a Keithley 2400 current source and HP 34420A nanovoltmeter. To measure cell performances, the fabricated solar

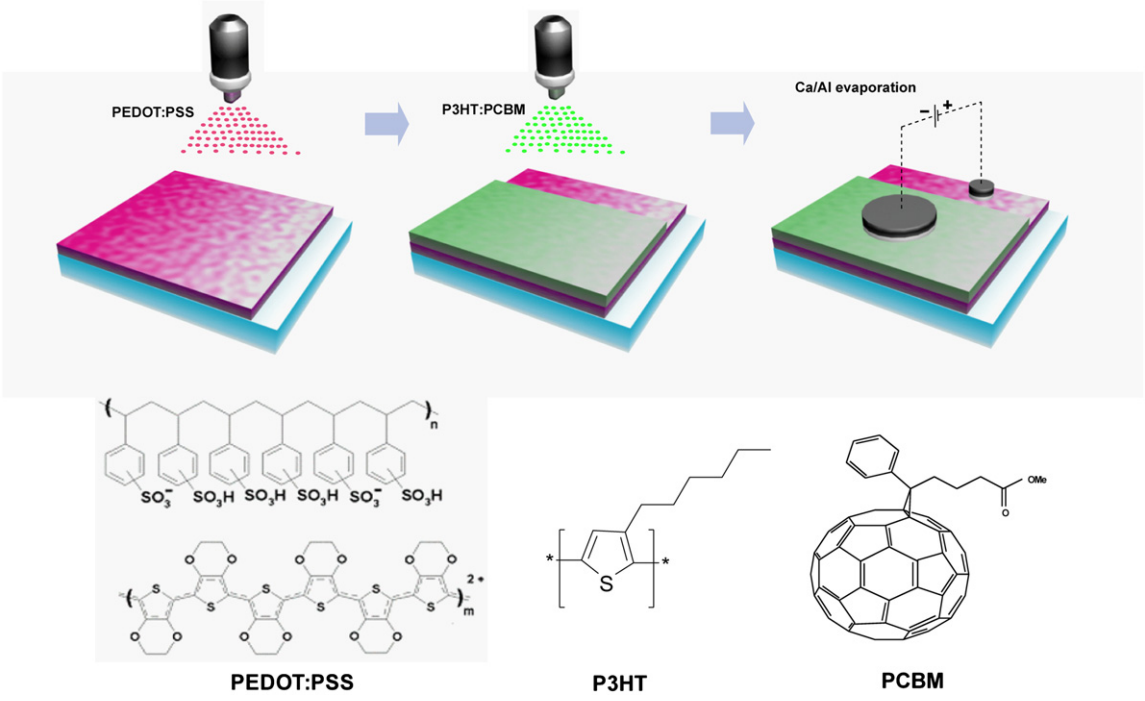

Fig. 1. Schematic representation of the fabrication sequence for ITO-free bulk-heterojunction solar cells with a spray-coated PEDOT:PSS anode and a spray-coated P3HT:PCBM active layer fabricated using a simple spray deposition (upper part) and chemical structures of materials used for the fabrication of ITO-free cells (lower part).

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cells were illuminated under 100 mW/cm^2 intensity generated from a 1 kW Oriel solar simulator with an AM 1.5 G filter in a N_2-filled glove box. The photocurrent density–voltage (J–V) measurements were performed using a Keithley 4200 source measurement unit. For accurate measurements, the light intensity was calibrated with a radiant power meter and a reference silicon solar cell (PVM188 with a KG5 color-filtered window) certified by the National Renewable Energy Laboratory (NREL).

3. Results and discussion

First, the optical transmission spectra of modified PH 500 films on glass substrates were investigated, as shown in Fig. 2a; the transmittances of a glass and a reference anode (ITO/4083) for conventional OSCs are also shown. The modified PH 500 films were spray-coated for either 2, 4, or 8 min of spray deposition time. As shown in Fig. 2a, compared with the reference anode, the PH 500 anode film sprayed for 2 min shows relatively good transparency in the full visible range, and as expected, the transmittance decreased as spray deposition time increased. Fig. 2b shows transmittance and sheet resistance as functions of the thickness of the spray-coated PH 500 films. The thicknesses of PH 500 anodes spray-coated at various deposition times were ~90 nm at 2 min, ~180 nm at 4 min, and ~340 nm at 8 min, respectively. The thicknesses of sprayed films were determined using optical effective thickness obtained from the UV–vis absorption spectra, because the sprayed films were too rough (the rms roughness of ~30 nm) to accurately measure their film thicknesses [25,30]. A wavelength range of 400–600 nm and a wavelength of ~500 nm were the main absorption region and the wavelength of maximum absorption of the photoactive P3HT, respectively. Thus, the transmittance was plotted for each PH 500 anode at a wavelength of 500 nm. The sheet resistance ($R_{sheet} = 1/\sigma \times d$, where σ is the conductivity, and d is the layer thickness) of each PEDOT:PSS film was evaluated using four-point-probe measurements. As shown in Fig. 2, the transmittance and sheet resistance were varied from 89.0% to 67.4% and from 358 to 63.3 Ω/squares, respectively, corresponding to an increase in film thickness. Although compared with the crystalline ITO (~10 Ω/square), most polymeric anode films have higher R_{sheet} and absorb more incident light, the properties of the spray-coated PH 500 films shown in Fig. 2 are considered to be sufficient for transparent and conductive electrodes in organic devices [37–39].

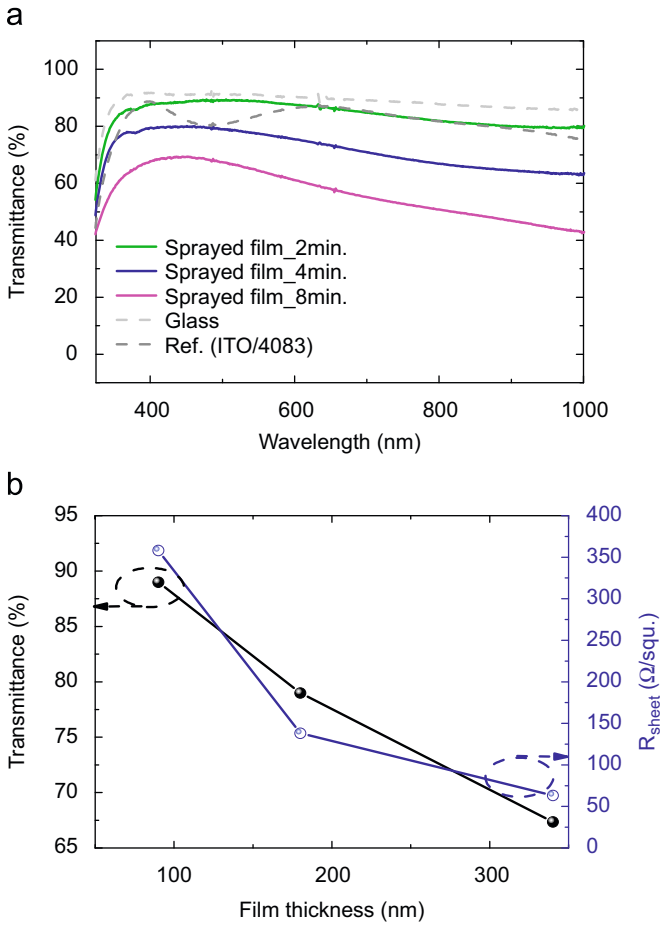

Fig. 2. (a) Optical transmission spectra of PH 500 films on glass substrates. (b) Transmittance at a wavelength of 500 nm and sheet resistance as a function of the thickness of the spray-coated PH 500 films.

To directly evaluate the fully spray-coated OSCs and investigate the effect of the thickness of PH 500 anodes on cell performance, we fabricated various solar cells comprising different PEDOT:PSS films spray-coated for 2, 4, 6, or 8 min and a subsequent spray-coated P3HT/PCBM layer. The performance of the fully spray-coated OSCs with different PEDOT:PSS films as anodes are compared in Fig. 3. As shown in Fig. 3a, fill factor (FF) values in cells with PEDOT:PSS anodes increased with spray deposition time. In general, the FF was tightly linked to the series resistance (R_s), and R_s is the sum of the bulk resistance and the contact resistance of the materials. Here, because all materials and processes used to fabricate the OSCs were identical except for the thickness of the PH 500 anodes, the R_s and FF of the OSCs can be dependent on the R_{sheet} of the PEDOT:PSS anodes. Therefore, it is believed that as spray deposition time increases, the increase in FF from ~34% to ~54% is attributed to a decrease in the R_{sheet} with increasing anode thickness, as shown in Fig. 2b. Although PH 500 anode film spray-coated for 8 min showed the maximum value of FF due to a decrease in R_{sheet} with increasing anode thickness, OSCs with the PH 500 anode film had the lowest short-circuit current density (J_{sc}), as shown in Fig. 3b. Considering that a thinner PEDOT:PSS film will conduct more incident light inside the photoactive layer and therefore will create more excitons, it is believed that the decrease of J_{sc} was due to the reduction of transmittance as film thickness increased, as shown in Fig. 2. It indicates that a trade-off exists between the transmittance and the series or sheet resistance, and thus, the PEDOT:PSS thickness should be carefully adjusted to balance them and optimized for cell efficiency. As a result, the fully spray-coated OSCs with the PH 500 anode that was spray-coated for 6 min showed the highest efficiency.

To investigate the feasibility of the fully spray-coated OSCs, ITO-free organic solar cell (IFOSC) with the PH 500 anode that was spray-coated for 6 min and two reference ITO-based OSCs using spin-coating a standard VPAI 4083 and the modified PH 500 solutions to be used as an interfacial layer in ITO-based cells were fabricated and compared. Performance characteristics of the three cells were obtained under 100 mW cm^{-2} illumination with AM 1.5G conditions and under the dark condition, respectively. As shown in Fig. 4, the performance characteristics of the IFOSC with the PH 500 anode were as follows: FF was 53.7%; open-circuit voltage (V_{oc}) was 0.61 V; J_{sc} was 6.62 mA/cm^2; and, the power conversion efficiency (PCE) was 2.17%. On the other hand, the performance characteristics of the two reference cells were obtained as follows: FF, 56.7% and 59.8%; V_{oc}, 0.59 and 0.59 V; J_{sc}, 7.98 and 8.06 mA/cm^2; and, PCE, 2.67% and 2.86%. These values are comparable with previous reports of devices with spray-coated photoactive layers [25,26]. Here, the slightly enhanced cell performance in the ITO-based cell with modified PH 500 as an interfacial layer was attributed to higher conductivity of the

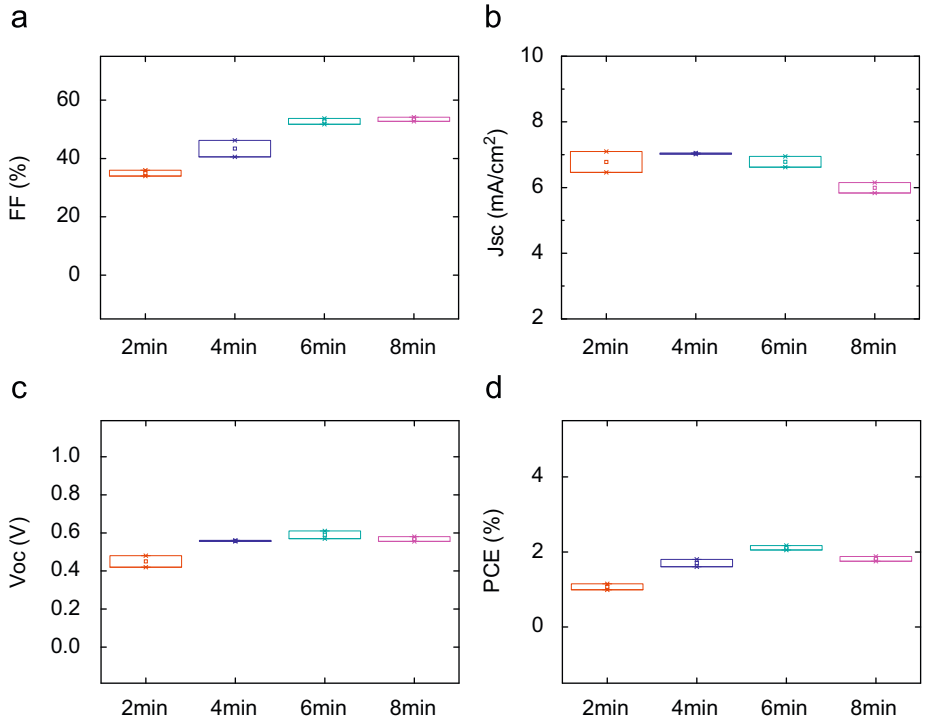

Fig. 3. Influence of the thickness of PH 500 anodes on the (a) fill factor (FF), (b) short-circuit current density (J_{sc}), (c) open-circuit voltage (V_{oc}), and (d) power conversion efficiency (PCE) of solar cells.

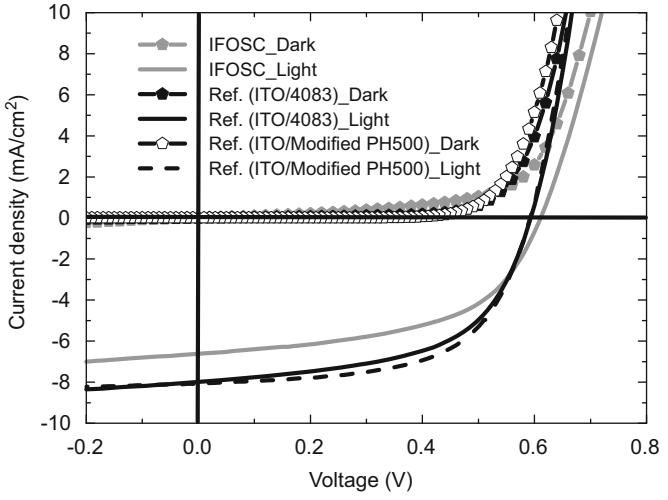

Fig. 4. *J–V* curves measured under illumination and in dark for three kinds of solar cells: a fully spray-coated ITO-free cell with the spray-coated PH 500 anode; an ITO-based solar cell with ITO/VPAI 4083 as a reference anode; and an ITO-based solar cell with ITO/modified PH 500 as a second reference anode.

modified PH 500 film than that of the VPAI 4083, which can reduce the series resistance in the ITO-based cells [40,41].

More importantly, as demonstrated in Fig. 4, the primary difference between the device with the PH 500 anode and those constructed with ITO is in FF and J_{sc}. Compared with the ITO-based cells, the decrease in FF and J_{sc} is believed to come from a higher series resistance in the IFOSC and a lower transmittance when using the PH 500 anode, which is confirmed by the dark curves shown in Fig. 4 and by the transmittance of the spray-coated PH 500 films shown in Fig. 2. Nevertheless, the overall photovoltaic characteristics of the cell with the spray-coated PH 500 anode were comparable to those of the ITO-based reference cell, indicating that the polymeric film has a similar function in the solar cell as ITO. In addition, the qualitative similarity of the photovoltaic characteristics shown in Fig. 4 indicates the feasibility of achieving all-spray-deposited OSCs without the conventional spin-coating method and the possibility of replacing the costly vacuum-deposited indium tin oxide (ITO) with highly conductive polymer films fabricated by inexpensive spray deposition techniques.

4. Conclusion

In conclusion, highly cost-effective ITO-free organic solar cells fabricated using a simple spray deposition method were demonstrated. All solution-processable layers of solar cells, highly conductive PEDOT:PSS anode films and photoactive P3HT:PCBM films, were spray-coated. PEDOT:PSS anode films with various thicknesses were prepared by controlling the spray deposition time. The transmittance and sheet resistance of PEDOT:PSS anodes were varied from 89.0% to 67.4% and from 358 to 63.3 Ω/squares, respectively, corresponding to an increase in film thickness. Fully spray-coated OSCs, including the spray-coated PEDOT:PSS anode layer and the spray-coated photoactive layer, presented a high power conversion efficiency of 2.17%. More importantly, the overall photovoltaic characteristics of fully spray-coated ITO-free OSCs were comparable to those of the ITO-based reference cell, demonstrating the feasibility of achieving all-spray-deposited OSCs without having to resort to use of the conventional spin-coating method. Furthermore, these results suggest the possibility of replacing ITO—prepared using cost-intensive vacuum processes and high temperatures—with highly conductive polymeric material prepared using simple spray deposition techniques. This fully spray-coated, ITO-free OSC will advance the realization of low-cost and high-efficiency polymer solar cells based on high-throughput roll-to-roll manufacturing.

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