# Fabrication of Spray-Coated Semitransparent Organic Solar Cells

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**ABSTRACT** We investigated a promising, low-cost method for fabrication of semitransparent organic solar cells by mass production. The active layer of the organic solar cells was added by spray coating with a dual action airbrush. The solution for the active layer was prepared from a rigorously blended poly(3-hexylthiophene-2,5-diyl) (P3HT) and (6,6)-Phenyl-C61 butyric acid methyl ester (PCBM) in 1,2-dichlorobenzene, and the surface morphology of the spray-coated active layer depending on the concentration of the P3HT and PCBM was investigated. The semitransparency achieved, came from the use of poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) as the conductive polymer electrode. For comparison, spin-coated solar cells were also fabricated. Power-conversion efficiency and transparency was achieved from the lower cost spray-coating method that was comparable to those by the traditional spin-coating method. The best spray-coated solar cell exhibited power-conversion efficiency of 1.9% (average or 1.7%) while the best spin-coated solar cell was 2.0% (average of 1.6%), when both were measured under the AM1.5G spectrum 100 mW/cm$^2$ light. Transmittance of the spray-coated solar cell was 52.2% while that of the spin-coated solar cell was 51.2%.

**INDEX TERMS** Spray-coating, organic solar cell, semitransparent, inverted, conductive polymer.

## I. INTRODUCTION

The installed cost of PV systems and corresponding levelized cost of electricity (LCOE) has rapidly declined and is expected to decline further. In Q1 of 2017, the installed price of a residential PV system was 2.66 USD/W$_{dc}$ (corresponding LCOE of 0.151 USD/kWh), and the National Renewable Energy Laboratory (NREL) has targeted the reduction of installed residential PV systems down to 1.1 USD/W$_{dc}$ (corresponding LCOE of 0.05 USD/kWh) by 2030 [1]. The price of a PV module accounts for only 13% of the installed PV system price; therefore, reduction of the PV module cost (USD/W) with high efficiency (high performance) is quite challenging due to the marginal efficiency premium [2]. Significant cost reduction may come from either the use of low cost materials or more economical fabrication methods.

In this paper, low cost organic solar cell fabrication methods via solution-processing techniques [3]–[6] were investigated. Options for solution-processing film deposition (coating) techniques include spin coating, blade coating, and spray coating. Spin coating [7] is the most widely used coating method and can easily be adjusted to the film thickness and provide an even surface. However, spin coating is very wasteful of the solution and potential for scaling to large area coating is limited. Blade coating (also known as knife coating) [8] is the simplest coating method. Blade coating can minimize the waste of solution, but provides a coarse film. Spray coating [9], [10] is a technique that is feasible for mass production, by which a solution is sprayed from a nozzle using compressed air. The merit to using spray coating is that it can cover large areas quickly and consumes only a small amount of solution. By adjusting the pressure

of the compressed air, the number of droplets and corresponding film thickness can be managed. Abdellah *et al.* separately deposited the P3HT and PCBM photoactive layer using alternate spray coating method and achieved the large interfacial area bulk heterojunction (BHJ) for efficient exciton dissociation [11]. However, in this paper, spray coating of rigorously blended active layer solutions were investigated for simple and efficient BHJ layer formation and final solution-processed organic solar cells.

In general, optimization of chemical composition and deposition condition of the BHJ active layer is to obtain bi-continuous and percolating morphology which is desirable for carrier transport. Du *et al.* [12] investigated the morphology of the BHJ active layer depending on the volume fraction of the P3HT and PCBM, and found that even volume fraction of P3HT and PCBM in solvent provided bi-continuous and percolating morphology. In this paper, morphology changes depending on the amount of the P3HT and PCBM in solvent, and deposition method (spray vs spin-coating) were investigated. A dual-action airbrush, which cost less than 100 USD, was used for spray coating the active layer of the organic solar cells. For the purpose of comparison, spin-coated solar cells were also fabricated. Then, the optical and electrical performance of the spray-coated solar cells was compared with that of the spin-coated solar cells.

## II. EXPERIMENTAL

Inverted-structure organic solar cells were fabricated [13], [14]. Fabrication of a solution-processed inverted organic solar cells started with indium-tin-oxide (ITO) deposited on a glass substrate. The substrate was then cleaned with acetone and isopropyl alcohol, followed by treatment with UV-Ozone. ITO worked as the cathode and the sheet resistance of the ITO was 10 $\Omega$/sq. On top of the ITO, zinc acetate solution was spin-coated to form a zinc oxide (ZnO) electron transport layer (ETL) [15], [16]. This was followed by spin-coating with polyethylenimine ethoxylated (PEIE) solution to form an interfacial buffer layer.

The solution for the BHJ active layer was composed of a mixture of 10∼20 mg poly(3-hexylthiophene-2,5-diyl) (P3HT, Lumtec LT-S909), 10∼20 mg (6,6)-Phenyl-C61 butyric acid methyl ester (PCBM, Lumtec LT-S905), and 1mL 1,2-dichlorobenzene. For the percolating and bi-continuous nanoscale morphology, wt% of P3HT and PCBM were even [12]. For spray-coating of BHJ active layer, a dual-action airbrush (BEETLE BUG, BBA-002) with round shape nozzle diameter of 0.2 mm was used. Single shot spraying was performed with air pressure of 1 bar and nozzle to sample distance of ∼20cm. For the counterpart samples, the active layer was formed from spin coating with a chuck speed of 400 rpm for 30 s. Both spray- and spin-coated samples were then annealed at 130 °C for 10 minutes. Note, only the active layer was either spray- or spin-coated, while all the other layers were spin-coated.
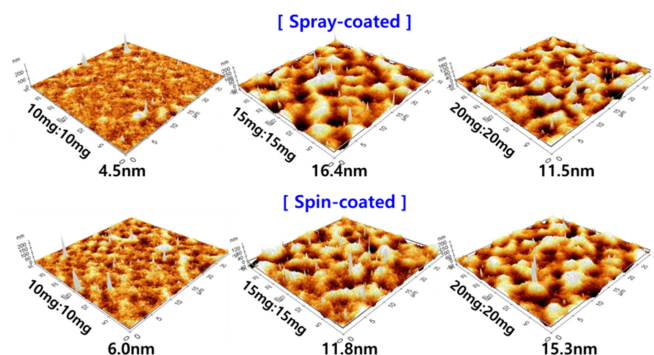


**FIGURE 1.** AFM image of the BHJ active layer from spray and spin coating.
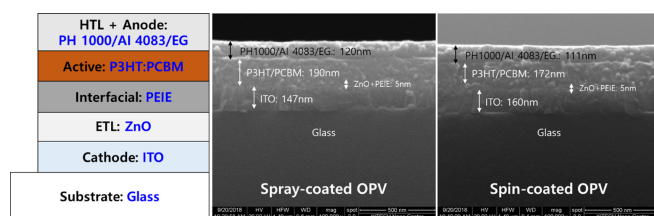


**FIGURE 2.** Geometrical structure and FE-SEM pictures of the fabricated spray- and spin-coated organic solar cells.

Finally, the hole-transport layer (HTL) and anode were made from a conductive polymer. The conductive polymer solution was a mixture of AI 4083 (Heraeus CLEVIOSTM P VP), PH 1000 (Heraeus CLEVIOSTM), PEDOT:PSS, and ethylene glycol (EG) [17], [18]. The solution was then spin-coated at 1000 rpm for 30 s followed by annealing at 130 °C for 3 minutes. Note, only solution processes were used to fabricate both spray- and spin-coated solar cells, and all processes were performed in normal air condition.

## III. RESULTS AND DISCUSSION

### A. SURFACE ROUGHNESS OF SPRAY-COATED VS SPIN-COATED ORGANIC SOLAR CELLS

Fig. 1 shows the atomic force microscopy (AFM) images of the spray-coated and spin-coated organic solar cells depending on the amount of the P3HT and PCBM in 1ml 1,2-dichlorobenzene. The numbers in the picture denote the amount of the P3HT and PCBM (XXmg:XXmg) in 1ml 1,2-dichlorobenzene and the rms roughness values in nm. As the concentration of the P3HT and PCBM increases, the rms roughness values are tend to increase due to the increased BHJ active layer thickness (Fig. 3). However, both spray- and spin-coated solar cells have comparable rms roughness values with comparable number of spikes. Comparable surface morphology between spray and spin coating may result comparable final device performance.

### B. FE-SEM PICTURE OF SPRAY-COATED VS SPIN-COATED ORGANIC SOLAR CELLS

Fig. 2 shows the geometrical structure and field-emission scanning electron microscope (FE-SEM) images of the fabricated solar cells with spray- or spin-coated active layer.
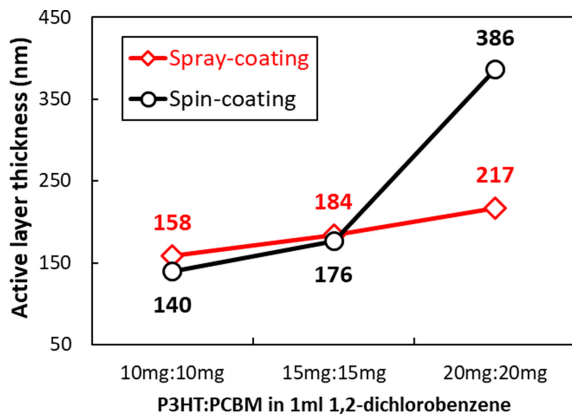
**FIGURE 3.** BHJ active layer thickness in relation to the amount of P3HT and PCBM in 1,2-dichlorobenzene, and to coating methods.

A solution of 15 mg P3HT and 15 mg PCBM in 1mL of 1,2-dichlorobenzene was spray-coated or spin-coated to form the BHJ active layer. Note, only the BHJ active layer was spray- or spin-coated, while all the other layers: ETL, interfacial layer, HTL, and anode, were spin-coated. The different coating methods resulted in different thickness of the active layer. This was 190 and 172 nm for spray- and spin-coated active layers, respectively, but there was no significant difference in the thickness of the other layers.

The thickness of the BHJ active layer can also be strongly determined by the concentration of organic materials (P3HT and PCBM). Therefore, the thickness of the BHJ active layer was measured in relation to the concentration of organic materials and coating methods. Fig. 3 shows the thickness of the BHJ active layer depending on the amount of P3HT and PCBM in the 1,2-dichlorobenzene and on the coating methods. Note that all spin coatings were performed at the rotation speed of 400 rpm for 30 s. It is interesting that, although the amount of P3HT:PCBM increased from 10:10 mg to 20:20 mg, there was no significant increase in the BHJ active-layer thickness from spray-coating while there was almost a three-times increase from spin coating. This is because solutions of higher concentration have higher viscosity and therefore result in greater thickness when spin coated at the same rotation speed, while spray coating does not strongly rely on the viscosity of the solution.

Because the BHJ active-layer thickness from spray coating is not strongly dependent on the viscosity of the solution, one can easily adjust the active layer thickness by increasing the number of spray shot to approach the optimum. This is because the lifetime of excitons generated occurs within a few-nanometer range in organic materials, so active-layer thickness far beyond exciton diffusion length is not desirable for collection of the generated photo carriers.

## C. TRANSMITTANCE OF SPRAY-COATED VS SPIN-COATED ORGANIC SOLAR CELLS

In this section, semitransparency of the fabricated final organic solar cells was investigated. Semitransparency was
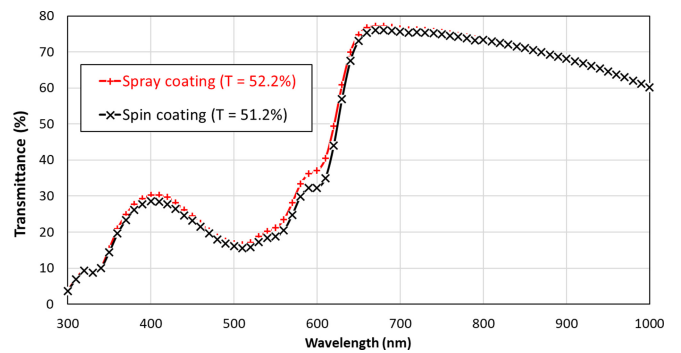


**FIGURE 4.** Transmittance of the fabricated final organic solar cells with spray- or spin-coated active layer.
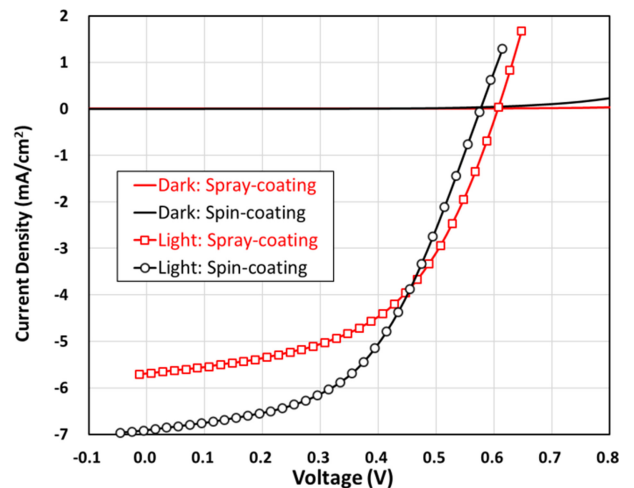


**FIGURE 5.** Dark and light I-V curve of the fabricated spray- and spin-coated solar cells.

achieved using a transparent conductive polymer electrode. Conductive polymer made of PEDOT:PSS (AI 4083 + PH 1000) and EG. Fig. 4 shows the transmittance curve of the final solar cell with a spray- or spin-coated active layer. The active-layer solution used in Fig. 4 was a mixture of 15 mg P3HT: 15 mg PCBM: 1 mL 1,2-dichlorobenzene. Both spray- and spin-coated solar cells showed a comparable transmittance value with average weighted solar transmittance (measured from 300 to 1200nm) of 52.2 and 51.2% for spray and spin coating, respectively. Comparable transmittance may come from the similarity in thickness of the layers of the spray- and spin-coated solar cells (Fig. 2).

## D. I-V CHARACTERISTICS OF THE SPRAY-COATED VS SPIN-COATED ORGANIC SOLAR CELLS

Fig. 5 shows the dark and light current-voltage (I-V) curve of the best solar cells fabricated with spray- or spin-coated active layer. The solid line represents the dark I-V curve, and marked line represents the AM1.5G spectrum (100 mW/cm$^2$) illuminated I-V curve. The reverse saturation current of the spray-coated solar cell was lower than that of the spin-coated solar cell. This could result in less leakage current, and as

**TABLE 1.** Solar cell parameters of the spray- and spin-coated solar cells.

| | | $V_{oc}$ (V) | $J_{sc}$ (mA/cm$^2$) | FF (%) | PCE (%) |
|---|---|---|---|---|---|
| | Best | 0.608 | 5.74 | 53.2 | 1.9 |
| Spray-coat. | Avg. | 0.601 | 5.39 | 52.3 | 1.7 |
| | Std. | 0.010 | 0.33 | 3.2 | 0.2 |
| | Best | 0.576 | 6.91 | 51.2 | 2.0 |
| Spin-coat. | Avg. | 0.581 | 6.49 | 41.4 | 1.6 |
| | Std. | 0.008 | 0.94 | 6.3 | 0.4 |

a result, a solar cell with spray-coated active layer provided higher open circuit voltage ($V_{oc}$) than did the solar cell with a spin-coated active layer. However, the spray-coated solar cell provided an inferior short circuit current ($J_{sc}$) and comparable FF. Inferior $J_{sc}$ may come from less uniform morphology of the BHJ active layer, and comparable FF comes from same electrode material (PEDOT:PSS). As a result, the spray-coated solar cell provided a comparable final PCE of 1.9% while the spin-coated solar cell PCE was 2.0%.

It is interesting that the average PCE value of the spray-coated solar cells was slightly higher (1.7%) than those of spin-coated solar cells (1.6%) due to the lower standard deviation. The detailed solar cell parameters are listed in Table 1. Note that the number of each spray- and spin-coated cells were around ten, and the area of a solar cell (active layer) was $2 \times 2$ mm, and the active layer of both solar cells was fabricated from a solution of 15 mg P3HT and 15 mg PCBM, in 1 mL of 1,2-dichlorobenzene.

## IV. CONCLUSION

Solution-processed semitransparent organic solar cells were fabricated by spray-coating and use of a conductive polymer electrode. A dual-action airbrush that could be bought in many stores was used for spray coating of the BHJ active layer. The surface morphology, optical and electrical performance of solar cells with spray-coated BHJ active layers was compared with that of solar cells with all spin-coated layers. Both spray- and spin-coated organic solar cells provided average solar weighted transmittance over 50%. In addition, when measured under AM1.5G (100 mW/cm$^2$) illumination, the spray-coated solar cell provided PCE of 1.9% (accounted for by the $V_{oc}$ of 608 mV, $J_{sc}$ of 5.74 mA/cm$^2$, and FF of 53.2%), while the spin-coated solar cell provided PCE of 2.0% (accounted for by the $V_{oc}$ of 576 mV, $J_{sc}$ of 6.91 mA/cm$^2$, and FF of 51.2%).

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