The effect of substrate temperature on the active layer for spray-deposition process in organic solar cells

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Abstract. In this paper, we fabricated organic solar cells using spray-coating with substrate heated method. We heated substrate temperature to 50 °C, 100 °C, and 150 °C respectively, during spray-coating process and observed different morphologies of the active layer. The difference of morphologies affected the performance of the organic solar cell. One of these samples, substrate heated as 150 °C, showed an open voltage of 0.61 V, short current density of 20 mA/cm² and power conversion efficiency of 3.84%.

Key words

Spray-coating, substrate heating, organic solar cell

1. Introduction

Shortage of energy by the exhaustion of the earth's natural resources made people look for new energy which can supply the demands for modern society. Solar energy which is sufficient to supply, harmless, free from pollution and having endless capacity is emerging alternative suggestion and regarded as an essential source of energy by many developed nations - use huge sum of investment via research facility - and try to make the material at a stage of practical use. So several kinds of solar cells are being developed[1]-[7]. Among them, there is a solar cell, which is processed via silicon, available as commercialized item with some advantages; considerable amount of efficiency of 20%, long life expectancy[1],[2]. Nevertheless, many research facilities strive to make a step further in developing 'organic solar cell'. Organic solar cell is regarded as an energy source that will replace the silicon based solar cell becouse it could be processed by solution process[1], [9]-[13]. The solution prcess is implementable to fabricate low cost and large-area structure by easy manufacture process like spin coating, ink-jet printing, knife-over-edge coating, slot-die coating and screen printing process[14]-[18]. One of those fabrication methodes, spray-coating method, compared with spin coating, consumes less solution and has no restriction to select substrate that have possibility to be flexible device and fabricated by roll-to-roll process[18]-[20].

In this study, we fabricated organic solar cells using spray-coating method in different substrate heating temperature of 50 °C, 100 °C, and 150 °C, and then we investigated the morphology of the active layers in each condition and measured performance of OSCs.

2. Experimental

In this study, the organic solar cells were fabricated through this process. The active solution to be formed photoactive layer by spray-coating method have conjugate polymer-fullerene derivative compounds, which is known as bulk heterojunction(BHJ) structure. donor, regioregular(rr) poly(3-hexylthiophene) As (P3HT; Sigma-Aldrich, St. Louis, MO) was used and phenyl-C71-butyric-acid-methylester (PCBM: ADS. Quebec, Canada) was used as acceptor, 30 mg/ml at weight ratio 1:1 in Dichlorobenzene(DCBZ), which was prepared in room temperature and stirred for 24 h. Indium tin oxide(ITO) patterned glasses(sheet resistance of below 20 ohm/square) of 30 mm by 30 mm were cleaned in acetone, methanol and deionized water, in that order. The cleaned glasses were blown with N₂ gas and dried at 150 °C for 10 min, after that modified Poly (3,4ethylenedioxythiophene)-polystyrene sulfonate (PEDOT:PSS; Clevios PH500) was spin-coated at 3000 rpm for 30 sec for form thickness of 24 nm on the cleaned ITO glass. Active layer was sprayed for 20 sec onto PEDOT:PSS layer annealed in 150 °C for 10 min, in conditions of the distance from spray nozzle to substrate was 20 cm and the sprayed pressure of N₂ was 0.1 MPa, when the substrate was heated to 50 °C, 100 °C, and 150 °C by hotplate respectively. Finally, Al was evaporated 150 nm as cathode on the active layer and post-annealed at 150 °C for 10 min. The active area of the fabricated organic solar cell was 0.09 cm².

The morphology roughness of active layer could be showed by using confocal laser scanning microscope (CLSM; Olympus, Tokyo, Japan), because it can display surface depending on the focus distance, and optical microscope also used. To measure the performance of the fabricated OSC, solar simulator was prepared in condition of AM 1.5 G, 100 mW/Cm² of xenon arc lamp which is calibrated by reference cell and filter, when I-V sourcemeter (Keithley 2400; Keithley Instruments Inc., Cleveland, OH) operated.

3. Result and discussion

We fabricated organic solar cells which were consisted of four layer, transparent electrode(ITO glass)/PEDOT:PSS/Photoactive layer/electrode(Al). For fabrication of photoactive layer, spray-coating deposition system was used. We established the spray-coating system to maintain the substrate temperature using the hot plate. The structure of the spray-coating system and fabricated organic solar cell are shown in Fig. 1. (a) and (b). The substrate was heated by hotplate and maintained temperature of 50 °C, 100 °C and 150 °C, when the active solution sprayed on to PEDOT:PSS layer. To confirm the effect of substrate temperature, we observed physical change of the active layer morphology using optical microscope and CLSM. According to the substrate temperature, different morphologies of active laver were obtained. One of them, fabricated at 150 °C, was shown in Fig. 2. Those image shown in Fig. 2 indicate that sprayed solution was crystallized rapidly when it contact with heated substrate (before the spraydeposition process, the substrate had already been heat during 1 min) and subsequent sprayed solution was stacked up on dried particle continuously. The stacking of solution particle with the results that pin-hole free morphology was obtained. And it means that the film was optically dense.

The average sizes of sprayed particle were affected by substrate temperature and we could confirm sizes of sprayed particle with optical microscope shown Fig. 2. The sizes of sprayed particle fabricated at 50 °C, 100 °C and 150 °C were 33.9 um, 24.1 um and 19.8 um, respectively. The particle of sprayed solution fabricated at 150 °C had the smallest size and highest dense in our samples. In contrast, in the case of the sample fabricated on 50 °C heated substrate, the size of sprayed particle was largest in our samples and the surface morphology was flat similar with that of the unheated sample. Because the substrate temperature was not sufficient to be crystallize rapidly. Through CLSM image, the average roughness of active layer morphology fabricated at 50 °C, 100 °C and 150 °C observed. The average roughness of active layer was 86 nm, 125 nm and 156 nm, respectively. The information of average roughness and particle size of fabricated are Table I. As the substrate temperature increase, the surface roughness became rough. Although these samples showed different roughness, average thickness of sprayed active layer was measured about 400 nm.

Table I. – Average roughness and particle size of fabricated sample in condition of substrate temperature of 50 °C, 100 °C and 150 °C.

	Average roughness	Particle size
50 °C	86 nm	33.9 um
100 °C	125 nm	24.1 um
150 °C	156 nm	19.8 um



Fig. 1. (a) Structure of spray deposition system, (b) Fabricated organic solar cell device.



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Fig. 2. Surface morphology images of (a) optical microscopy and (b)CLSM. Spray deposition at 150 °C.

After observation of changed active layer surface morphology, we fabricated Al cathode. The performance of completed organic solar cell is shown Fig. 3. It shows J-V curve of fabricated organic solar cells. When the substrate was heated temperature of 50 °C, 100 °C and 150 °C, V_{OC} increased in order. However, J_{SC} maintained high current density, about 20 mA/cm². Because we used modified PEDOT:PSS material, which is mixture of pure PEDOT:PSS and 5% dimethyl formamide(DMF). Enhancement of electric conductivity of polymer with adding solvent is researched by J.Y. Kim et al[21]. That is the main reason of the high current density.

Because increased surface roughness which was affected by substrate temperature helped contact with a electrode and photoactive layer when those devices post annealed. However, in spite of high current density of 20 mA/cm², we obtained low efficiency. This result came from low fill factor that was obtained about 0.3 for all samples fabricated at 50 °C, 100 °C and 150 °C. In conclusion, the power conversion efficiencies(PCE) of OSCs were improved 3.03%, 3.26% and 3.84% according to increased V_{oc}.



Fig. 3. J-V curves of spray deposited OSCs with different substrate temperature.

4. Conclusion

We demonstrated the effect of substrate temperatrue during spray deposition process. We fabricated OSCs using spray deposition method in different substrate temperature of 50 °C, 100 °C and 150 °C during spray coating process. To investigate the effect of substrate temperature, we observed the morphology and roughness of active layer. The substrate temperature affected surface morphologies of active layer and performance of the organic solar cell. In conclusion, when we measured the performance of the device fabricated at 150 °C showed open circuit voltage of 0.61 V, short circuit current density of 20 mA/cm² and PCE of 3.84%. The PCE of this device was low for high current density. It was considered that the PCE was affected by low fill factor. The fill factor depends on mobility, lifetime of carrier and thickness of the active layer. Also it is sensitive parameter with oxygen in the air[22]. In here, the low fill factor was attributed that P3HT/ PCBM blending solution particles were oxidized during spray deposition process. This degradation could be reduced by making in N₂ atmosphere and we can get improved OSCs with high PCE and low cost process.

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