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Scalable Fabrication of Flexible Large-area Inverted Organic Photovoltaic Cells

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***Abstract*—Scalable large-area inverted organic photovoltaic (OPV) cells were fabricated on a flexible polyethylene naphthalate (PEN) substrate. To account for the scalability and impact of the substrate (rigid versus flexible), OPVs of various sizes (0.04–1.6 cm^2) were fabricated on flexible PEN and rigid glass substrates. A single OPV cell area of up to 1.6 cm^2 can be fabricated using a solution process at a low temperature of < 160 ℃. It should be noted that all processes except those of the electrodes were conducted based on a solution process under ambient air conditions, not inside a N_2 filled glove box. It was found from the numerical calculations and experimental measurements that a lower photoconversion efficiency (PCE) for higher areas mainly comes from a degradation in the fill factor (FF). Solar cell characteristic parameters were measured under an AM1.5G spectrum (intensity of 100 mW/cm^2), and the PCE was 1.8% and 2.0% for the OPV on the PEN and glass substrates, respectively.**

***Index Terms*—Large-area OPV, flexible substrate, solution-process, low-temperature process, scalable**

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I. INTRODUCTION

The installation of photovoltaic (PV) systems has been increasing rapidly owing to a rapid reduction in the cost of PV panels. Because panel costs are considered in dollars per watt, either a low cost or high performance can be beneficial [1, 2]. Among the many low-cost PV technologies available, organic photovoltaics (OPVs) are attracting attention owing to their low fabrication cost, printability, and simple device structure. A low fabrication cost could come not only from smaller facility costs but also a shorter fabrication time and a low thermal budget. In addition, the intrinsic nature of polymer organic materials is compatible with a flexible substrate. The portable, deformable, and stretchable nature of this flexible OPV can be used for foldable or wearable devices. A single-junction OPV achieves its best PCE of 17.35% when the area is 0.032 cm^2 [3]. However, the best PCE of single-junction OPVs with an area of 1.023 and 26.129 cm^2 has decreased to 13.45% and 12.6%, respectively [3]. Furthermore, the PCE of the best OPV module with an area of 802 cm^2 was 8.7% [3].

Many institutes have also investigated large-scale OPVs on top of flexible substrates. Han et al. reported an all-solution process OPV on a flexible polyethyleneterephthalate (PET) substrate with a PCE of 5.25% with an area of 80 cm^2 [4]. Krebs et al. reported a roll-to-roll processed (R2R) OPV on a flexible PET substrate with a PCE of 2.1% for an active area of 120 cm^2 [5]. Strohm et al. also reported a R2R processed 60 cm^2 PCE of a 5% OPV module [6].

In a previous study, a large-scale OPV was achieved from the series connection of each small-scale cell. However, in this research, we focused on the fabrication

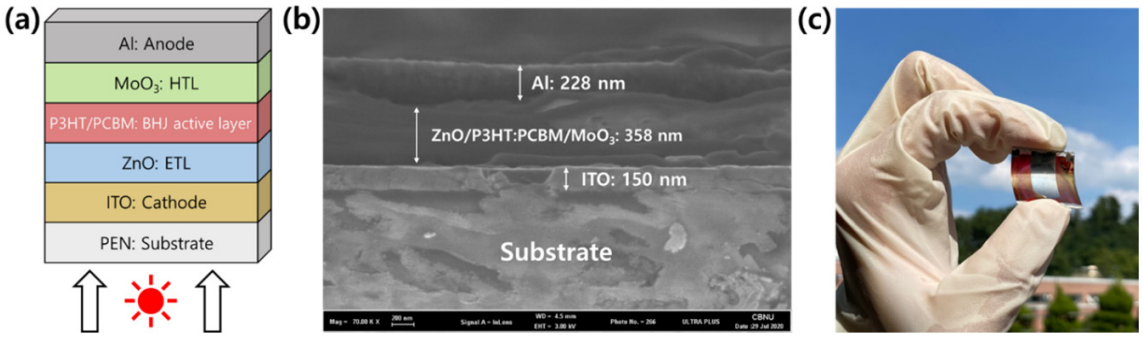

Fig. 1. (a) Geometrical structure, (b) FE-SEM, (c) photograph of the fabricated flexible inverted OPV.

of large single cells using feasible low-cost mass-production PV fabrication methods such as low-temperature (low thermal budget), vacuum-free (time budget), and solution-based (low facility cost) processes [7, 8]. Various OPV sizes of 0.041–1.6 cm^2 were fabricated from a low-cost solution process for a systematic study of the loss analysis of large-scale OPVs. In addition, numerical calculations from experimental data provide a roadmap for the performance and loss of large-scale OPVs.

II. Experimental

The fabrication of a flexible large-area inverted solution-processed OPV starts from an indium tin oxide (ITO, 15 Ω/sq) deposited polyethylene naphthalate (PEN) substrate (thickness of 125 μm) followed by the patterning of ITO for a specific designated area. ITO patterning was conducted using a 1:1 volume ratio of deionized water to hydrochloric acid for 20 min, and a cathode electrode was applied for an inverted OPV structure [9]. After ITO patterning, substrates were cleaned in a sonicator with acetone and isopropyl alcohol for 10 min. Subsequently, the ZnO layer was added to the ITO as an electron transport layer (ETL). ZnO is formed from the spin-coating of a solution containing 1.6 g of zinc acetate (Alfa Aesar, 97%) in 0.4 g of ethanolamine and 66 g of ethanol at 2000 rpm for 30 s followed by annealing at 160 °C on a hot plate for 25 min [10, 11]. On top of the ZnO ETL layer, a 0.1 wt% polyethylenimine ethoxylated (PEIE) solution was spin-coated at 4500 rpm for 30 s and then annealed at 80 ℃ for 10 min for an interfacial buffer layer.

Subsequently, bulk heterojunction (BHJ) photoactive layers were formed from a mixed solution of 1:1 poly (3-hexylthiophene-2, 5-diyl) (P3HT, Lumtec LT-S909) and (6, 6)-Phenyl-C61 butyric acid methyl ester (PCBM, Lumtec LT-S905) in 1,2-dichlorobenzene (99% Sigma Aldrich). This solution was then spin-coated at 400 rpm for 30 s followed by annealing at 110 °C for 10 min. A slightly low spin speed of 400 rpm is required for a thicker active layer to prevent pin-hole induced shunting, which may significantly degrade the solar cell performance for large areas. Finally, a hole transport layer (HTL) and an anode metal electrode were deposited from an ultra-high vacuum (10^{-7} torr) thermal evaporator (Selcos CETUS OL 100). A total of 1 nm of MoO_3 and 200 nm of Al were deposited as an HTL and an anode electrode, respectively. For the reference (rigid OPV) device, all of the above processes were applied equally to a 0.7-mm thick glass substrate. Solar cell characteristic parameters were measured using a Class A AM 1.5 G spectrum solar simulator (Wacom WXS-155S-L2).

III. Results and Discussion

1. Geometrical Structure of Fabricated Inverted OPVs

Fig. 1 shows the (a) geometrical structure, (b) field emission scanning electron microscopy (FE-SEM, Zeiss Ultra Plus), and (c) developed solution-processed large-area inverted OPVs on a flexible substrate (PEN). The inverted structure has an anode electrode on top and a cathode electrode on the bottom. To determine the feasibility of low-cost mass production, the reason for the adoption of the inverted structure was to fabricate OPVs under ambient air conditions, not inside a N_2 filled glove box. This is because inverted OPVs have been reported for high stability under air ambient than conventional OPVs [12]. Moreover, as HTL and anode electrode are

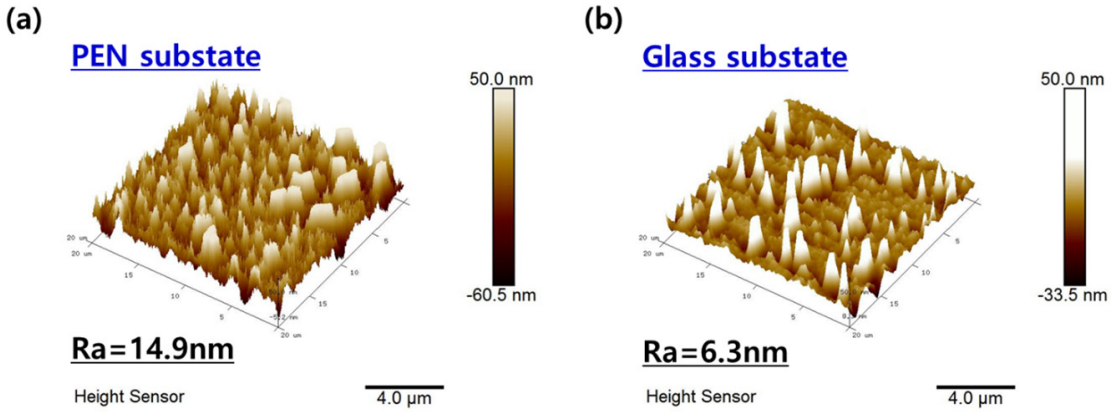

Fig. 2. AFM images of fabricated final OPV with (a) PEN, (b) glass substrates.

intimate in inverted OPV, so realization of all-solution process could be possible by replacing HTL and anode electrode to aqueous materials such as conductive polymer [7, 8].

The thicknesses of the cathode and anode were 150 and 228 nm, respectively, with semiconducting materials having a thickness of 358 nm. It should be noted that all processes except the anode electrode were applied from a solution with a low temperature of below 160 °C, which also opens the possibility of a low thermal budget production, such as a roll-to-roll process, for this type of OPV fabrication. Note that the sample for the FE-SEM image was fabricated onto a rigid glass substrate because of the clear FE-SEM image.

Fig. 2 shows an atomic force microscopy (AFM, Bruker Dimension Icon) image of the final OPVs fabricated on glass and PEN substrates. The average roughness (Ra) of the OPV with a PEN substrate was 13.8 nm, whereas that with a glass substrate was 6.3 nm. As expected, the Ra of the OPV with the PEN substrate was higher because the rigid substrate induced an even spread of the liquid solution and easy handling.

2. Characteristic Parameters of Scalable Flexible Large-area Inverted Organic Photovoltaic Cells

This section evaluates the scalability of the developed solution-process flexible inverted OPVs. In addition to the PCE, the area of the solar cell is critical for its power production. One technique that cannot scale-up up to a PV module of a certain area cannot be chosen by the customer. Therefore, in this section, OPVs with different areas (0.04–1.6 cm^2) were fabricated on glass and PEN substrates to account for the impact of the area of the OPV and the rigidity of the substrate. Fig. 3 shows the normalized (a) open-circuit voltage (V_{oc}), (b) short-circuit current density (J_{sc}), (c) fill factor (FF), and (d) photoconversion efficiency (PCE) of the OPV with a glass or PEN substrate as a function of its area. It was found that V_{oc} has no significant difference for a different solar cell area. A slight degradation of the J_{sc} may come from the less even spread of the solution materials for larger areas owing to the limitations of the spin-coating.

A significant degradation for higher area solar cells was mainly due to FF degradation. The FF of the solar cell can be defined as Eq. (1). Where FF_o is the ideal fill factors which does not account for parasitic resistance, v_{oc} is a normalized voltage calculated from the measured V_{oc}, and r_s is a normalized resistance calculated from the measured R_{se} (series resistance), V_{oc}, and J_{sc} [13]. The R_{se} is mainly dependent on the sheet resistance (R_{sh}) of the anode and cathode electrodes and the area of the solar cell, as shown in Eq. (2) [14]. As the thickness of the organic layer (t_{org}) was few hundred nanometers, so other series resistance components $\left(\rho_{org} \cdot t_{org} + r_{int}\right)$ can be negligible. Therefore, R_{se} will be almost double when the area is double and will degrade the corresponding FF of the solar cells. It should be noted that ρ_{org} and t_{org} are the resistivity and thickness of the organic bulk materials, respectively, r_{int} is the interface resistance. From the numerical calculations, more 1cm^2 area degrades absolute FF of ~11% and ~15% for OPV with glass and PEN substrates. As determined experimentally, an

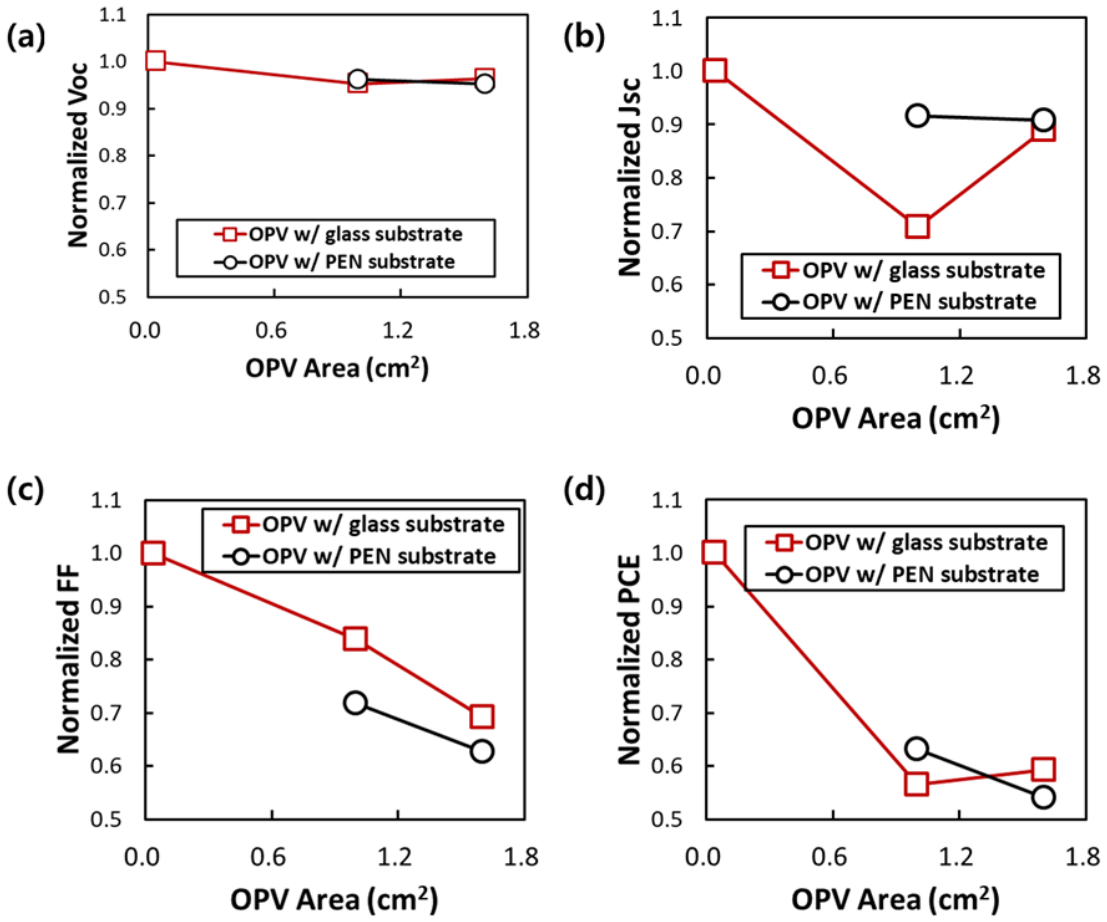

Fig. 3. Normalized solar cell characteristic parameters (a) V_{oc}, (b) J_{sc}, (c) FF, (d) PCE as a function of the solar cell area.

absolute FF of 10.1% occurred as the area of the solar cell increased from 0.04 to 1.0 cm^2.

$$FF = FF_o\left(1-r_s\right),\ FF_o = \frac{v_{oc} - ln\left(v_{oc}+0.72\right)}{v_{oc}+1},$$
$$v_{oc} = \frac{qV_{oc}}{nkT},\ r_s = \frac{R_{se}}{V_{oc}/J_{sc}} \quad (1)$$

$$R_{se} = R_{sh}\left(\frac{L}{W}\right)Area + \sum_N\left(\rho_{org}\cdot t_{org} + r_{int}\right) \quad (2)$$

Fig. 4 shows the calculated FF as a function of the solar cell area with experimental data. Numerical calculations were conducted using Eqs. (1) and (2) with input from the experimental data. It can be seen clearly that the numerical calculations matched the experimental data for both the OPV with glass and PEN substrates. Fig. 4 also provides a road map for the solar cell performance for various areas. The performance of a solar cell when scaling up from its initial performance can be anticipated.

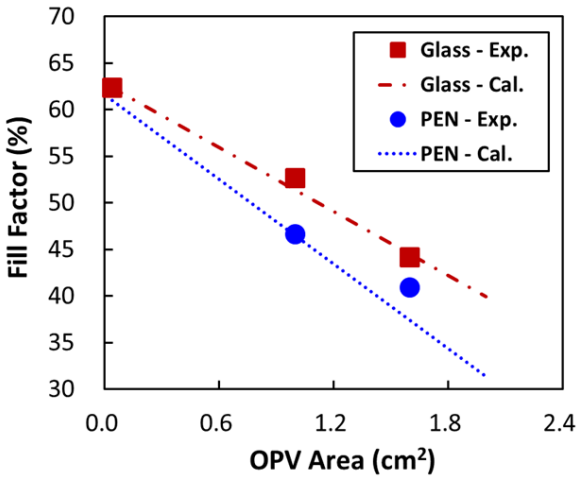

Fig. 4. Calculated and experimental fill factor values as a function of the solar cell area when fabricated on both glass and PEN substrates.

For example, if OPV area is increased from 1.0 to 3.0cm^2, FF will degrade from 52% to 30% and corresponding PCE will degrade from 2.0% to 1.2%.

Table 1. Solar cell characteristic parameters for fabricated best OPV for different areas with glass and PEN substrates

Substrate	Area (cm^2)	V_{oc} (mV)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)	R_{se} (Ω-cm^2)	R_{sh} (Ω-cm^2)
Glass	1.6	601	7.5	44.0	2.0	34.6	533
PEN	1.0	604	7.7	43.2	2.0	25.9	357
	1.6	599	7.6	38.9	1.8	41.5	314

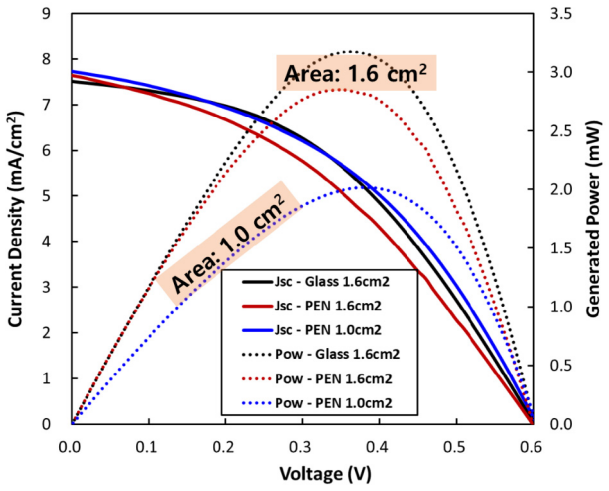

Fig. 5. I-V curve of the fabricated best OPV for different areas with glass and PEN substrates.

Table 1 summarizes the characteristic parameters of a solar cell, and Fig. 5 shows the I-V curve of the best OPV with different areas and substrates. As the area of the OPV increased from 1.0 to 1.6 cm^2, the FF decreased from 43.2% to 38.9%, and the corresponding PCE decreased from 2.0% to 1.8%, for the PEN substrate. For reference, OPV on a glass substrate with an area of 1.6 cm^2 provided an FF of 44.0% and a PCE of 2.0%. From this result, we can conclude that a PCE degradation for higher areas is mostly from an FF degradation. Therefore, from the numerical calculations in Fig. 4, we can achieve an FF of 31.3% and PCE of 1.4% when the area increased to 2.0 cm^2. In addition, as shown in Fig. 5, the generated power of the higher area produced a higher energy proportional to its area.

IV. Conclusion

Scalable inverted OPV cells were fabricated on a flexible PEN substrate. Single OPV cells with an area of over 1.6 cm^2 were fabricated using a solution process at a low temperature of below 160 °C. To quantify the PCE loss for large-area solar cells and the impact of the substrate, OPVs with various areas were fabricated using a flexible PEN and rigid glass substrates. The PCE degradation of both glass and PEN substrates showed a similar trend, whereas OPV with a PEN substrate showed a slightly inferior performance. From the numerical calculations using an experimental data input, the solution-processed OPV fabrication method on a flexible substrate could have a scalability of up to an area of 2.0 cm^2 without a significant decrease in the PCE. The PCE will be 2.0%, 1.8%, and 1.4% when the area of a single OPV is 1.0, 1.6, and 2.0 cm^2, respectively.

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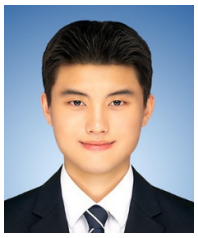

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