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Partitioning of the organic layers for the fabrication of high efficiency organic photovoltaic devices

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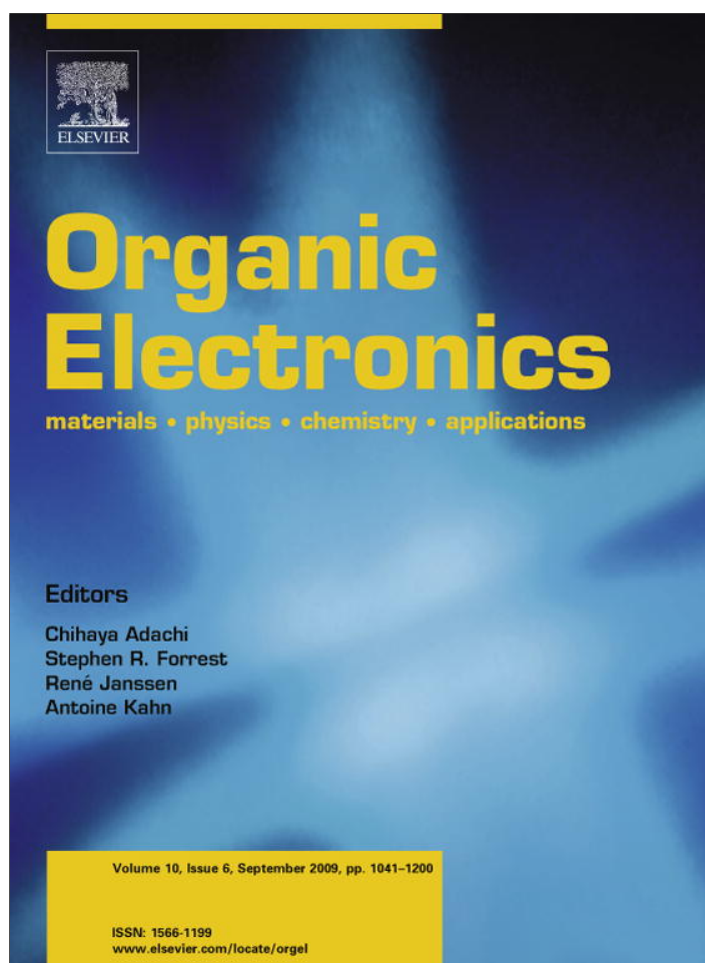
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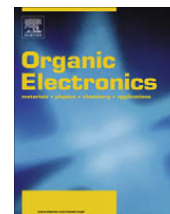
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ABSTRACT

Lateral partitioning of hole extraction layer with insulating walls improved the power conversion efficiency of organic photovoltaic device. When the conductivity of the hole extraction layer is low, no improvement is obtained by partitioning. However, when the conductivity is high, a significant improvement was obtained in the partitioned cells, showing the estimated power conversion efficiency of 4.58% compared to the 3.54% of the single cell structure. This improvement, carefully corrected by masking at measurement, could be explained by the reduction of series resistance. Although accurate estimation of device area at partitioned device might be difficult, its effectiveness on the properties of large area organic photovoltaic device is clear, as shown in the result of 1 cm²-size cell with the same manner.

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1. Introduction

Organic photovoltaic (OPV) device entails several advantages over traditional silicon-based or other inorganic solar cells, which includes possibility of custom tailoring structure, light weight, inexpensive and reproducible processability. Thus, OPV is accepted as a new way of obtaining clean renewable energy. Recent rapid improvements of the power conversion efficiency elicit anticipation for commercial OPV devices in the near future. The advent of bulk heterojunction interpenetrating network structure using polymer blends alleviates inherent

disadvantage of low conductivity [1]. Problems of narrow optical absorption are greatly suppressed with tandem cell structure with different absorption maximum semiconductors [2]. Increased current density for parallel connection or open circuit voltage for series connection [3] increases the efficiency, and the synthesis of low band gap materials removes solar emission mismatching problems [4]. However, the conversion efficiency of single OPV device is still far from a commercially viable value, which is estimated as about 10%, thus requiring further improvements in this direction. The conversion efficiency of OPV devices hitherto attainable is still not close to the approximate maximum value of 11% [5].

Most of polymer thin film photovoltaic devices are fabricated using polyalkylthiophenes/modified fullerenes

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blend on transparent indium tin oxide (ITO) glass. Such a polymer–organic blend system can be easily applied by simple spin coating or printing techniques. In order to achieve the maximum power conversion efficiency for a given material system, organic photovoltaic devices require low series resistance and high shunt resistance. It is also required to have two ohmic contacts in order that bulk series resistance is minimized; first is that between the positive electrode and the active layer, and second is that between the active layer and the metallic electrode. Sandwiching the active layer made of poly(3-hexylthiophene)/[6,6]-phenyl-C61-butyric acid methyl ester (P3HT/PCBM) blend is the hole extraction layer (poly(3,4-ethylenedioxythiophene):poly(4-styrene sulfonate) [PEDOT:PSS] coated ITO) and a negative electrode, metallic conductor such as

Al. Because of a relatively low conversion efficiency of single OPV devices, they need to be fabricated with large scale connectivity. On the other hand, resistivity at large area hole extraction and active layer will induce an increase of series resistance which results in a reduction of current flow, thus affecting the conversion efficiency. A close relationship between the device area and the conversion efficiency is now well established [6].

In this study, we focus on our efforts to partition the hole extraction layer into smaller areas by means of the insulating walls so as to minimize the lateral contribution on the series resistance, thus to prevent lowering of the efficiency by increasing the overall active area. We demonstrate that this method is universally applicable to all organic materials used in OPV. Also, our method will provide a distinct advantage of implementing by simple printing such as inkjet, gravure, offset and screen, lithography, and soft contact transfer. We should note here that our method differs from the conventional module fabrication technique that inserts low resistance metal lines between active areas.

2. Material and methods

2.1. Fabrication of devices

PEDOT:PSS (poly(3,4-ethylene dioxythiophene):poly(styrene sulfonate)) aqueous dispersions (Baytron® Al4083, P and PH-500) were used as the hole extraction layer, spin coated on top of 300–400 nm thick ITO coated

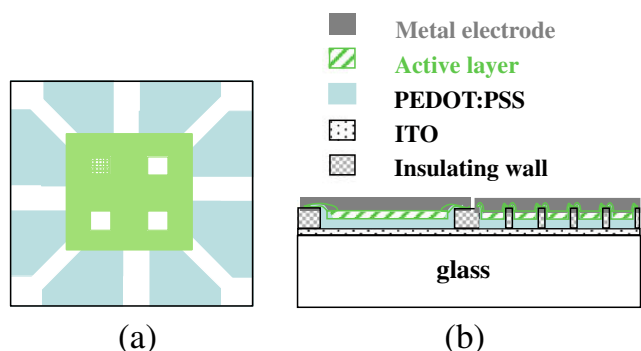


Fig. 1. (a) Schematics and actual picture of insulation layer coating on etched ITO and (b) cross-sectional schematics of fully processed device.

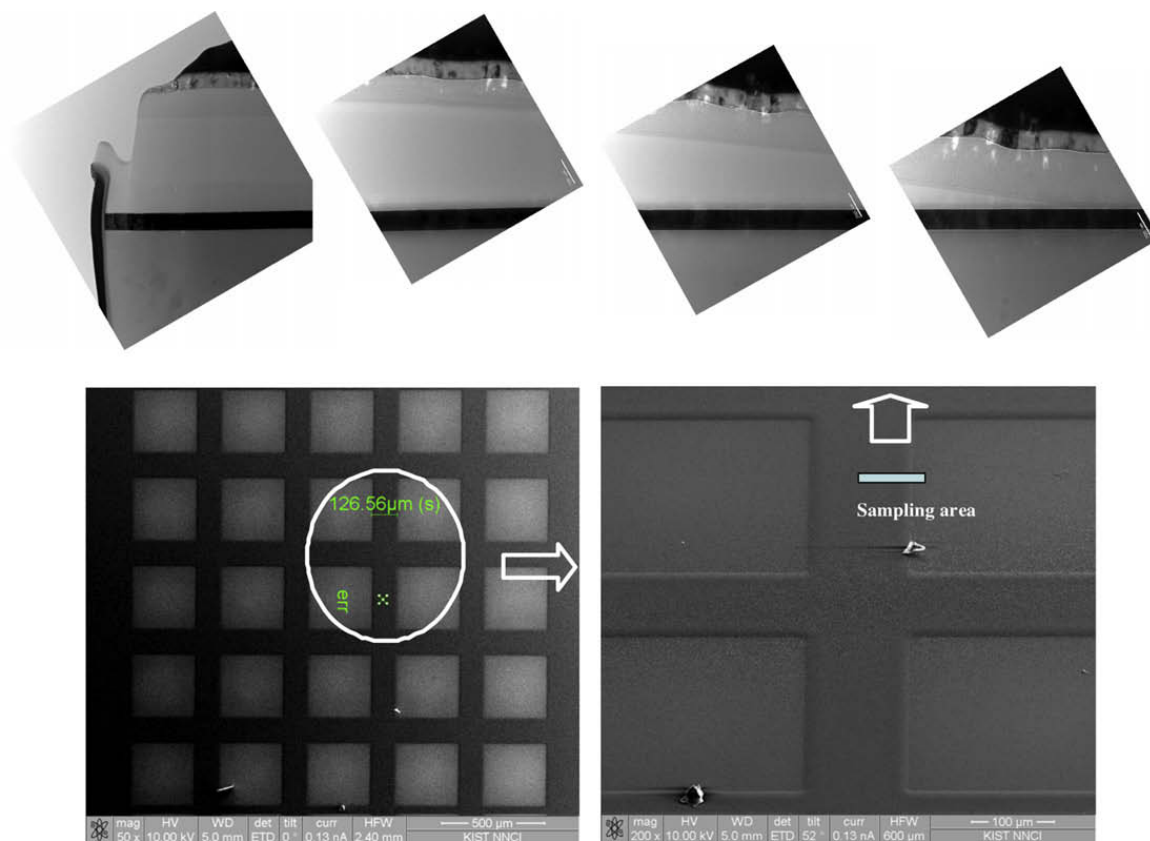


Fig. 2. FIB-TEM images; bottom left: partitioned cell, top images: cross-sectional images from insulating wall edge to center of wall.

glass. In the case of DMSO blended PH-500, the ratios of PH-500: methanol: DMSO were 1: 2: 5%, respectively. The PEDOT:PSS stock solution was spin coated at 2000 rpm for 40 seconds to form about 35 nm thick film, then was baked at 200 °C for 10 min in a high purity nitrogen environment glove box. ITO glass was cleaned by boiling in chloroform, isopropyl alcohol and acetone for 30 min in each solvent, sonification for 15 min in 50:50 isopropyl alcohol and acetone solution, and finally rinsed with deionized water. A typical example of the next step of sample preparation is as follows: 2.4 wt% of P3HT and PCBM blend with 1:0.6 ratio by weight dissolved in anhydrous chlorobenzene and was spin coated on prepared PEDOT:PSS coated ITO glass at 2500 rpm for 40 s. The thickness of resulting active layer was 220 nm and pre-annealed at 150 °C for 10 min. 0.8 nm thick LiF, which served as a buffer layer, was deposited on top of the active layer and 150 nm thick aluminum was evaporated by thermal vapor deposition at 10^{-6} – 10^{-7} Torr, and prepared device was post annealed at 120 °C for 10 min. All of the processing was performed in the glove box with high purity nitrogen envi-

ronment. Photocrosslinkable positive photoresist resin of acrylate derivatives (Nippon Zeon Corp.) was used to form partitioning wall of height of 500 μm . Standard partitioned subcell area was $300 \times 300 \mu\text{m}$, and the partitioning wall was 200 μm in width and 500 μm in height. After finishing the series of the preparation processes, the device was encapsulated with glass plate using an UV curable resin in a high purity nitrogen environment glove box.

2.2. Characterization

For cross-sectional transmission electron microscope (TEM) (FEI TECNAI F20) observation of device, a thin flake in situ lift-out sample was prepared using focused ion-beam (FIB) machine (FEI Nova600) with a Ga ion focused beam. Thickness of coated film was measured with surface profiler (TENCOR[®], P-10 α -step). Oriel Class A type solar simulator (IEC 904) with Oriel Reference Cell (calibrated data taken by NREL) was used as a light source and all measurement was performed under 1 sun condition ($100 \text{ mW}/\text{cm}^2$). The measurements were not corrected for reflection

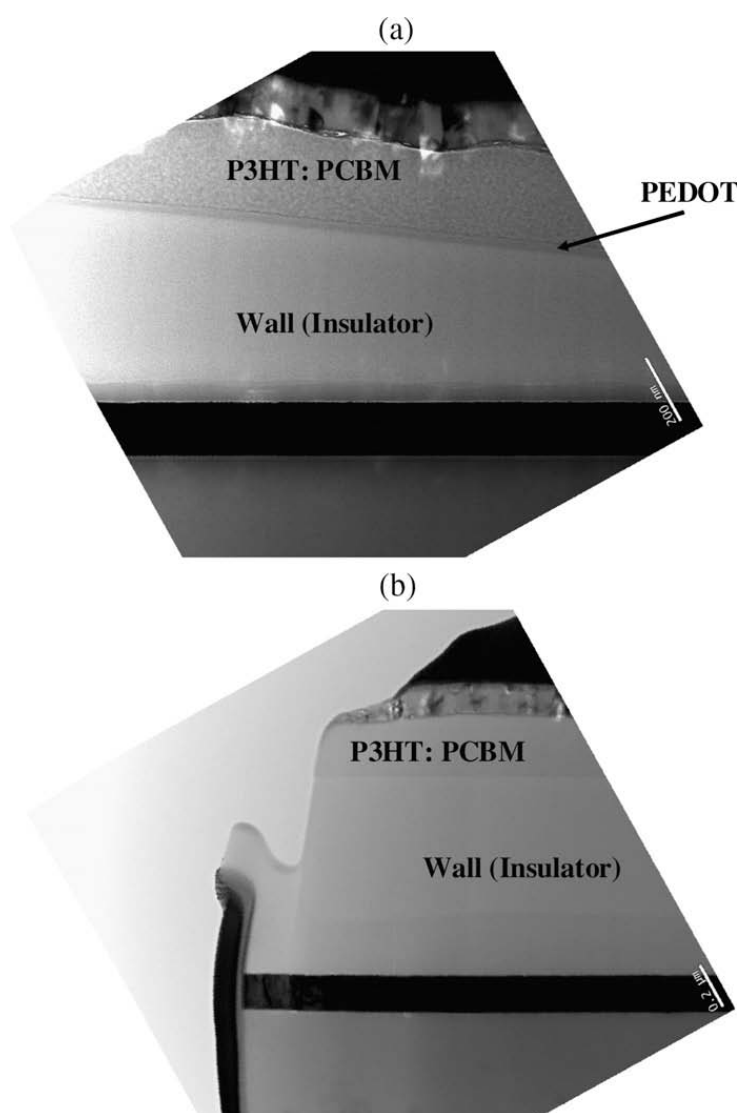


Fig. 3. FIB-TEM images; (a) interface area of partitioning wall and (b) center position of partitioning wall.

losses and light absorption in the ITO electrode. Current–voltage characteristics were determined with Keithley 2400 source–measure unit.

3. Results and discussion

In Fig. 1 is displayed our fabrication design of photovoltaic cells consisting of smaller sized subcells to the conventional cell size, i.e. 0.04 cm^2 . Eventually we have increased the total cell size to 1 cm^2 , to which we will return at the end. Parenthetically, the size of 1 cm^2 is quite a large cell for OPV. Our test coupon is made of three single cells of 0.04 cm^2 , one partitioned cell with actual active area of 0.027 cm^2 which consisting of 25 subcells of the same size ($300 \mu\text{m} \times 300 \mu\text{m}$), and $200 \mu\text{m}$ space between them. These are so formed by an insulator on top of one inch by one inch ITO coated glass as shown in Fig. 1. The partitioned subcells form a single cell upon covering its top with an Al electrode.

It is quite simple to build the insulating walls to disconnect the hole extraction layer by means of a curable resin via a lithographic method. With TEM micrographs in Fig. 2 and Fig. 3, we show that the hole extraction layer (PEDOT:PSS) is discontinued at few tens of micrometer away from the edge of insulating walls so that the series resistance is reduced through a smaller active area. Each divided active area acts like an isolated small cell so that the series resistance is reduced. In order to elucidate the effect of partitioning the active areas, a cell without partitioning wall is used as the reference.

Fig. 4 shows the profiles of current density vs. bias voltage (J – V) between single and partition cells for three different formulations of hole extraction layer. The results are

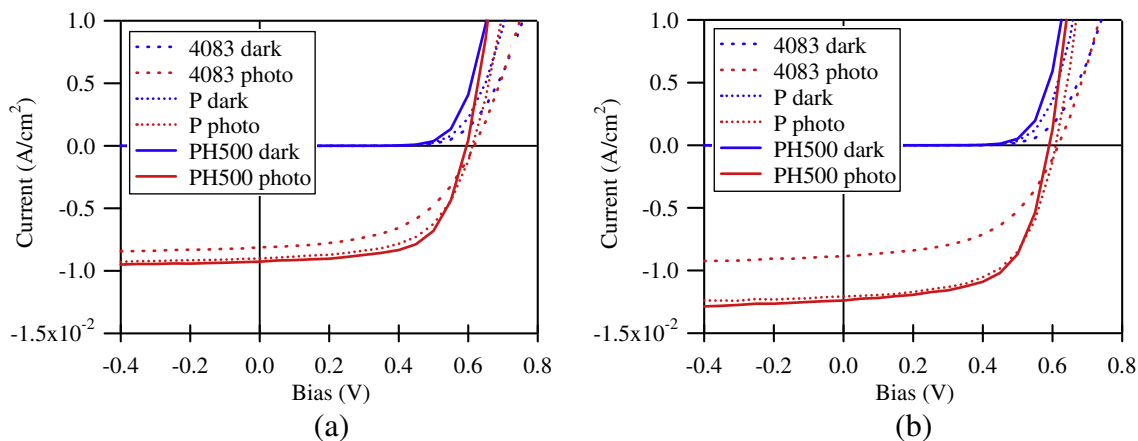


Fig. 4. J – V characteristics for three different hole extraction layers; (a) single cell and (b) partition cells.

Table 1

Influence of conductivity of hole extraction layer on performance of OPV device (*s*: single area cell; *p*: partitioned cell).

Conductive layer	Area	Open circuit voltage (V)	Current density (mA/cm^2)	Fill factor	Series resistance ($\Omega \text{ cm}^2$)	Efficiency
Baytron Al 4083 (10–3 S/cm)	<i>s</i>	0.617	8.12	0.526	7.47	2.64
	<i>p</i>	0.616	8.86	0.527	7.40	2.88
Baytron P (1 S/cm)	<i>s</i>	0.615	8.72	0.609	3.33	3.27
	<i>p</i>	0.609	12.06	0.602	2.43	4.42
Baytron PH-500 (100 S/cm)	<i>s</i>	0.596	9.27	0.641	2.22	3.54
	<i>p</i>	0.591	12.39	0.626	1.81	4.58

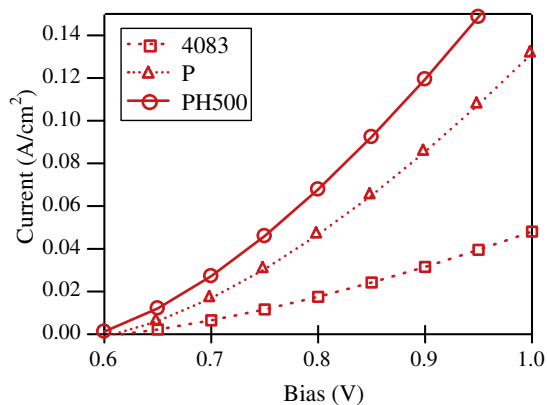


Fig. 5. Transmittance of ITO and insulating walls, and absorption characteristics of P3HT/PCBM blend.

summarized in Table 1. When Baytron 4083 is used as the hole extraction layer having a low conductivity (~ 10 – 3 S/cm), there was no detectable difference between the single and partitioned cells. However, when the conductivities were higher (Baytron P: $\sim 1 \text{ S/cm}$ and Baytron PH-500 blended with dimethyl sulfoxide: $\sim 100 \text{ S/cm}$), there were significant differences in the efficiency between the two, and the degree of this difference was in the order of conductivity of the hole extraction layer formulations. Henceforth, we refer Baytron PH-500 blended with dimethyl sulfoxide (DMSO) as simply PH-500. Clearly, the observed enhancements of the current density and efficiency are closely correlated to the conductivity of the hole extraction layer. Thus, we infer that there exist lateral contributions of the series resistance from the rest of active area to a subcell for the bulk resistance. As is shown in

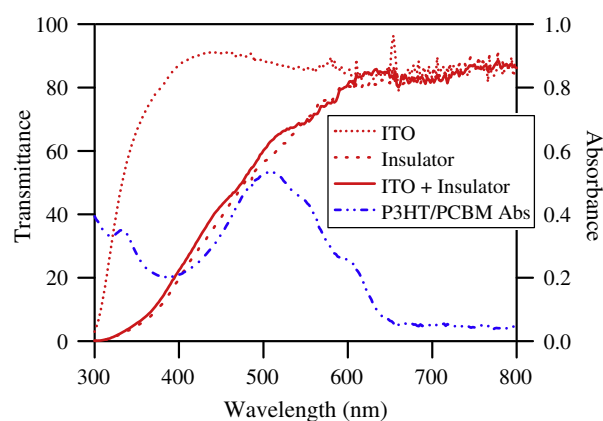
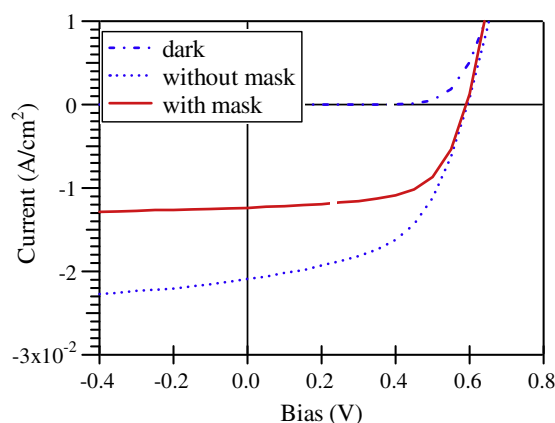
Table 2Influence of wall to active area ratio (active area was $300 \mu\text{m} \times 300 \mu\text{m}$).

Conductive layer	Wall width (μm)	Open circuit voltage (V)	Current density (mA/cm^2)	Fill factor	Series resistance (Ωcm^2)	Efficiency
Baytron Al 4083 ($10^{-3} \text{ S}/\text{cm}$)	None	0.590	8.53	0.460	22.68	2.35
	20	0.595	8.23	0.460	25.62	2.27
	50	0.593	8.27	0.480	23.25	2.39
	100	0.595	8.47	0.490	15.49	2.46
	200	0.595	8.27	0.500	10.78	2.49
Baytron PH-500 ($100 \text{ S}/\text{cm}$)	None	0.596	8.09	0.570	8.09	2.75
	20	0.602	7.75	0.564	8.51	2.63
	50	0.592	9.23	0.586	6.21	3.20
	100	0.586	10.26	0.517	6.27	3.11
	200	0.596	12.93	0.561	4.63	4.32

Fig. 5, the lowering of series resistance can be observed clearly by the increasing slope of the current density vs. bias voltage profile above the open circuit voltage; the inverse of slope in the figure represents the bulk series resistance. Despite the enhanced conductivity of the hole extraction layer, it still has a relatively high resistance compared to that of the electrode. Such a high resistance of the hole extraction layer is attributed to the contributions from far away areas, whereas such is not the case with a low conductive hole extraction layer. In short, for highly conductive hole extraction layers, the deleterious effect of the lateral contributions of the series resistance from the rest of active areas to the bulk resistance is rather significant. Therefore, the enhancement of the efficiency will be substantial over that of a cell made with low conductivity formulations if the hole extraction layer is disconnected from each other by partitioning into smaller area subcells.

We have also investigated the influence of wall to active area ratio using different partitioned cells and the results are summarized in Table 2. The power conversion efficiency was increased with increasing partitioning wall width. When Baytron Al 4083 was used as the hole extraction layer, there was no detectable difference in the power conversion efficiency among the different wall widths. However, when Baytron PH-500 was used, there were significant differences in the efficiency and the series resistance among wall widths, and the degree of this difference was in the order of wall widths. This result is quite reasonable since lateral contributions of the series resistance from the rest of active area to the subcell should be decreased with increasing wall width.

We now turn to the uncertainty with respect to the estimate of active cell area. As was shown in Fig. 2 with the cross-sectional TEM micrographs by focused ion-beam (FIB), we estimate that the boundary of PEDOT:PSS coating is about $30 \mu\text{m}$ from the edge of insulating wall. Since PEDOT:PSS has to be coated over rather tall insulating walls, having a height of $500 \mu\text{m}$ at the center position, our cell coating process is different from the coating on flat ITO surface. Unlike the reported edge effect (effective areas are $300\text{--}400 \mu\text{m}$ from the edge) [7] when PEDOT:PSS is coated on a flat surface, the actual active area in our device starts about $30 \mu\text{m}$ away from the edge since the PEDOT:PSS does not extend flush to the edge. This issue is being raised because the size of active area is critical to the efficiency, particularly when the active area is small

**Fig. 6.** Transmittance of ITO and insulating walls, and absorption characteristics of P3HT/PCBM blend.**Fig. 7.** *J*-*V* characteristics of a partitioned device with and without mask.

[7]. In addition, the insulating wall materials with the $500 \mu\text{m}$ height at the center, as referred to in the above, absorb visible light and the transmittance at the P3HT/PCBM blend absorption region is less than 50% as shown in Fig. 6. Therefore, the actual exciton formation in the area between the insulating wall to the PEDOT:PSS coating edge may be small but still needs to be accounted for in the estimate of the efficiency. Thus, the actual active area in the partitioned cell is subject to a certain range of uncertainty. Nevertheless, we emphasize that the trend of increasing current extraction with a high conductivity of the hole extraction layer by partitioning is firmly established.

Table 3

Characteristics of large area cell.

Conductive layer	Area	Open circuit voltage (V)	Current density (mA/cm ²)	Fill factor	Series resistance (Ω cm ²)	Efficiency
Baytron Al 4083 (10 ⁻³ S/cm)	s	0.590	8.53	0.460	22.68	2.35
	p	0.595	8.27	0.500	10.78	2.49
Baytron PH-500 (100 S/cm)	s	0.596	8.09	0.570	8.09	2.75
	p	0.596	12.93	0.561	4.63	4.32

It is well known that artifacts caused by the edge effect are to be taken into account with care [7,8]. In fact, it is a point of a recent controversy with the solar cell efficiency [9]. According to the report by Cravino et al. [7] a small area cell like ours is significantly affected by the edge effect as expected. We thus examine the edge effect by masking the device area. Fig. 7 shows J - V characteristics of the partitioned device with and without mask. When the entire fabricated device was exposed to incident light without masking, the power conversion efficiency was about 6.5; the current density was ~ 20.9 mA/cm² and the fill factor was 0.523. On the other hand, when the mask was placed so as to expose only active area to the incident light, the power conversion efficiency was reduced to 4.6; the current density was ~ 12.4 mA/cm² and the fill factor was 0.626. Thus, we conclude that the edge effect is indeed significant relative to the actual power conversion efficiency. It can be readily understood that the current density decrease and fill factor increase are caused by the charges formed only in the active area.

Finally, we come to the issue of a large area size cell of 1 cm². Despite difficulty in quantifying the power conversion efficiency arising from the uncertainty of actual active area within the partitioned walls, the key point of this study is clear: One can fabricate a large area OPV device without sacrificing the power conversion efficiency, if not enhancing, when hole extraction layer is partitioned into smaller subcells. A large area cell (~ 1 cm²) was prepared by the same manner as described in the above. To elucidate the partitioning effect, a single area cell with the comparable active area was also prepared. The results are summarized in Table 3. As was seen in Table 3, the partitioned cell performance with Baytron Al 4083 showed no distinct difference from that of single cell (efficiency change from 2.35 to 2.49). On the other hand, the partitioned cell performance with Baytron PH-500 showed about 40% improvement in the efficiency over that of single cell (efficiency change from 2.75 to 4.32). Notwithstanding any uncertainty relative to the absolute estimate of the power conversion efficiency arising from the active area uncertainty, the results in Table 3 make it compelling that the partitioning works when compare to a device with the same overall active area.

Major factor causing this difference is the series resistance, noted by the slope of J - V curve beyond open circuit voltage (Fig. 5). As discussed previously, the partitioning of hole extraction layer caused the series resistance decrease. This is entirely consistent with a report by Yoo et al. [10] that the series resistance increased with the number of cells upon connecting them in series. Since the areas of these two cells were same, it was clear that a partitioned

cell with a high conductivity hole extraction layer would have a lower series resistance resulting in enhanced current extraction leading to improved power conversion efficiency.

4. Conclusion

We conclude with these results that the partitioning of the active area into smaller areas can improve the power conversion efficiency of organic photovoltaic devices. The lateral serial contributions to the series bulk resistance of the hole extraction layer with a high conductivity profoundly affect the power conversion efficiency. Our method reported here is to be differentiated from the conventional metal line insertion for the purposes of increasing the current output as used in photovoltaic industry.

Acknowledgments

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