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Improved Performance in Polymer-Fullerene Blend Photovoltaic Cells by Insertion of C60 Interlayer

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We demonstrate the enhancement of the device performance in polymer:PCBM blend photovoltaic cells by insertion of C60 interlayer between the active layer and the upper electrode. The active layer was fabricated by spin coating of a polymer [(poly(2-methoxy-5-(2'-ethyl-hexyloxy)-1,4-phenylene vinylene)(MEH-PPV) or (poly(3-hexylthiophene)(P3HT) and fullerene derivative [5,6]-phenyl-C61-butyric acid methyl ester(PCBM)), then the C60 interlayer and LiF/Al electrode were deposited. The both polymer cells with C60 interlayer showed significant increase in the power conversion efficiency compared to cells without the C60 layer.

Keywords: buffer layer; bulk-heterojunction; organic photovoltaics; polymer photovoltaics

1. INTRODUCTION

Organic photovoltaic cells (OPCs) are attracting much interest due to some potential advantages of low weight, mechanical flexibility, and low cost production by use of printing technologies. The bulk heterojunction (BHJ) type OPCs, which have the interdigitated bicontinuous mixture of an electron donor and an acceptor, have been reported to

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show relatively high quantum generation efficiency of charge carriers [1]. Uchida *et al.*, demonstrated the significantly improved performance in a small molecule-based OPC with an organic p-i-n structure which has molecularly mixed BHJ layer sandwiched between a donor layer and an acceptor layer [2–4]. Organic conjugated polymers such as poly(phenylene-vinylene) [PPV] derivatives and polythiophene derivatives, have been also applied for OPCs. As the recent developments in synthetic methods and device fabrication technologies, the polymer PCs have continued to be improved in the power conversion efficiency (η_p) [5,6]. Until 2006, over 5% of η_p have been reported [7]. They employed the BHJ consisting of a conjugated polymer and a fullerene derivative, (6,6)-phenyl-C61-butyric acid methyl ester (PCBM). The carriers generated at the donor/acceptor interface were transported through the polymer network and the micro-segregated PCBM cluster. To improve the carrier collection efficiency at the electrodes in the BHJ cell, it is critically important to design the electrode/active layer structure carefully. Lots of studies have been reported to increase the efficiency by optimizing the upper electrode [8] and inserting a buffer layer [9].

In the present study, we demonstrate the enhancement of the device performance in polymer:PCBM blend photovoltaic cells by insertion of C60 interlayer between the active layer and the upper electrode. The cells with C60 interlayer showed significant increase in the η_p compared to cells without C60 layer.

2. EXPERIMENTAL DETAILS

2.1. Materials

The chemical structures of the used materials in this study are shown in Figure 1. The polymers, poly((2-methoxy-5-(2'-ethylhexyloxy)-1,

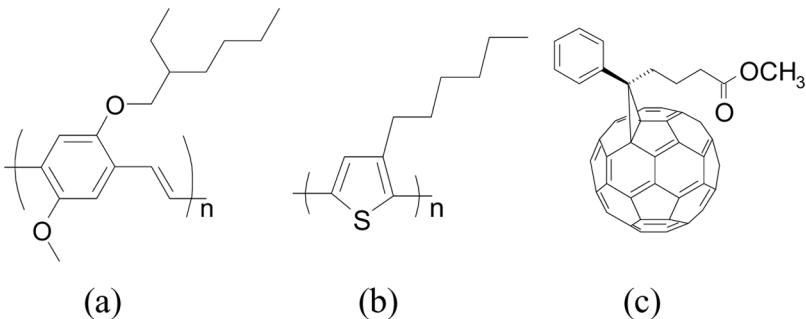


FIGURE 1 Chemical structures of MEH-PPV (a), and P3HT (b), and PCBM (c).

4-phenylenevinylene) (MEH-PPV) and poly(3-hexylthiophene) (P3HT) were purchased from American Dye Source and Aldrich, respectively. MEH-PPV was purified by reprecipitation twice from methanol before use. P3HT was used without further purification. The PCBM was synthesized as described in a literature [10] and used as an electron acceptor. A buffer layer material used poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate)(PEDOT:PSS)(Baytron 4083).

2.2. Fabrication of Blend Photovoltaic Cells

Figure 2 shows the configuration of the fabricated devices, indium tin oxide(ITO)/PEDOT:PSS/polymer:PCBM/LiF/Al, and inserting a C60 interlayer between the active layer and the upper electrode.

A pre-cleaned ITO substrate was coated with PEDOT:PSS, as a buffer layer, by spin-coating at 1500 rpm for 30 s, followed by 5 min drying at 120°C. The thickness of PEDOT layer was evaluated to be ~ 40 nm by a probe profiler, DEK-TAK3 (Ulvac). The chlorobenzene solutions at the concentration of 1.0 wt% of polymer:PCBM at the ratio of MEH-PPV:PCBM = 1:4, P3HT:PCBM = 2:1 were spun on the substrate. The film was then annealed at 150°C, 5 min. A C60 interlayer and LiF/Al cathode were deposited by a vacuum evaporator to make four kinds of devices with the active area of 2×2 mm². The device structures of A ~ G are shown in Figure 2.

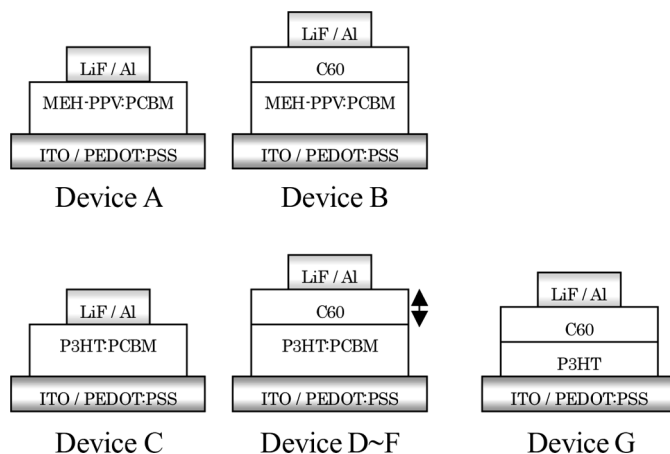


FIGURE 2 Configuration of the fabricated **Device A ~ F**. **Device A**: MEH-PPV:PCBM = 1:4, **Device B**: MEH-PPV:PCBM = 1:4/C60(10 nm), **Device C**: P3HT:PCBM = 2:1, **Device D ~ F**: P3HT:PCBM = 2:1/C60(5,10,30 nm), **Device G**: P3HT/C60.

For the measurements of the photocurrent action spectra, an incident light of Xenon lamp passing through a monochromator was irradiated onto the device from the ITO electrode side without applied bias. The current–voltage characteristics of the devices were measured with a Keithley 238 source measurement unit under conditions of both dark and illumination of 100 mW/cm^2 white light provided from a 1.5 AM solar simulator in the air ambient at room temperature.

3. RESULTS AND DISCUSSION

Figure 3a shows the J-V characteristic of Device A and Device B. The short circuit current density (J_{sc}) was significantly increased by inserting the C60 interlayer. Also in the case of P3HT:PCBM = 2:1 blend photovoltaic cells, the insertion of C60 interlayer (5 nm) caused the improvement of the cell performance (Fig. 3b). The PC characteristics of Device A ~ D are summarized in Table 1. Under the irradiation of AM1.5 at 100 mW/cm^2 , J_{sc} and η_p were 6.79 mA/cm^2 and 2.52% in Device A and those were increased to 7.70 mA/cm^2 to 2.82% in Device B. Also in the case of P3HT:PCBM, J_{sc} and η_p were increased 5.36 mA/cm^2 to 7.05 mA/cm^2 and 1.81% to 2.06% by the insertion of C60 interlayer.

Figure 4 shows the incident photon-to-current conversion efficiency (IPCE) spectrum of P3HT:PCBM devices with and without the C60 interlayer ($\sim 10 \text{ nm}$). The IPCE is defined as the number of photogenerated charge carriers contributing to the photocurrent per incident photon. Device C shows the typical spectral response of P3HT:PCBM [9–11] with a maximum IPCE of $\sim 50\%$ at around 500 nm. In

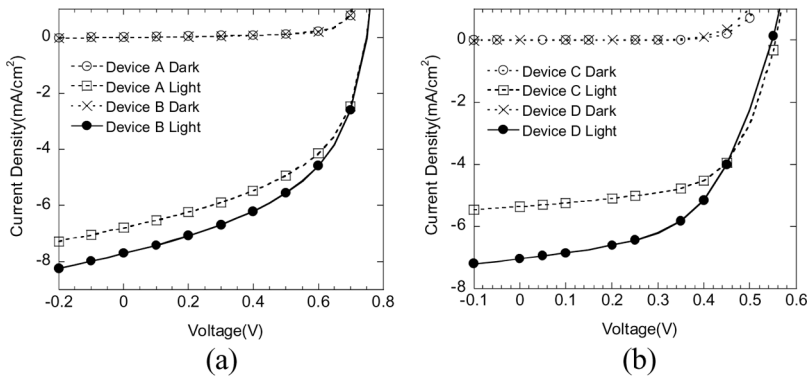


FIGURE 3 Current density-voltage characteristic of device A and B (a). And of device C and D (b).

TABLE 1 Photovoltaic Properties of Devices for Different Materials used in the BHJ Cells

Structure of BHJ	V_{oc} (V)	J_{sc} (mA/cm ²)	FF	η_p (%)
Device A C60 (0 nm)	0.75	6.79	0.49	2.52
Device B C60 (10 nm)	0.75	7.70	0.49	2.82
Device C C60 (0 nm)	0.56	5.36	0.60	1.81
Device D C60 (5 nm)	0.55	7.05	0.54	2.06

Device E, the IPCE was 30% higher than that of Device C at around the absorption range of P3HT due to the C60 interlayer (10 nm) insertion. In the varied C60 interlayer thickness from 0 to 30 nm. J_{sc} reached a maximum at 10 nm. (Table 2)

One of the reasons for the performance improvement by the insetion of C60 interlayer would be that the interface of BHJ and C60 can act as an additional donor-acceptor junction, i.e., exposed fraction of the polymer among PCBM on BHJ surface would contact to the deposited C60. However, this contribution would not be dominant, because Device G which has donor-acceptor (D-A) heterojunction (HJ) structure (ITO/PEDOT/P3HT/C60/LiF/Al) showed much smaller J_{sc} (0.8 mA/cm²) than the increases of J_{sc} by the C60 insertion, $\Delta J_{sc} \sim 2.0$ mA/cm², which is estimated from the J_{sc} difference between Device C and Device E. Since the contact area between the polymer and the C60 in Device E should be smaller than that in Device G due to the PCBM also occupying the surface on the BHJ.

Another reason of the improvement, which seems to be dominant, would be that the C60 layer blocks the holes to prevent the collection

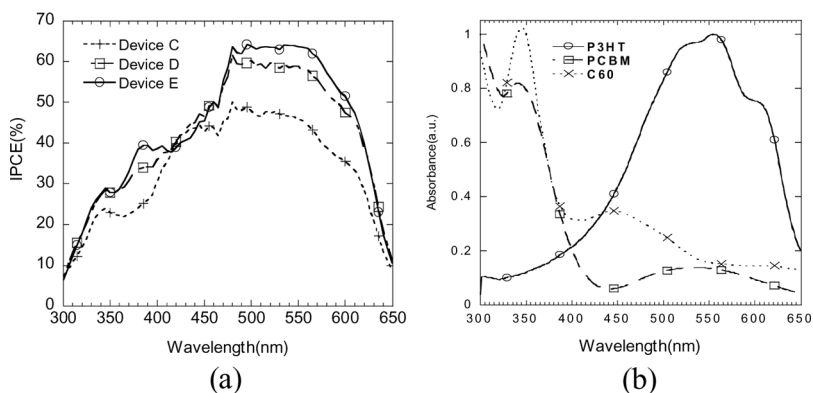
**FIGURE 4** IPCE spectra of device C~E (a) and UV-Vis spectra of P3HT, PCBM, C60 (b).

TABLE 2 Photovoltaic Properties of P3HT:PCBM = 2:1 Activelayer BHJ Devices for Dependence of the C60 Interlayer Thickness (Device C~F)

Structure of BHJ	V_{oc} (V)	J_{sc} (mA/cm ²)	FF	η_p (%)
Device C C60 (0 nm)	0.56	5.36	0.60	1.81
Device D C60 (5 nm)	0.55	7.05	0.54	2.06
Device E C60 (10 nm)	0.53	7.23	0.43	1.61
Device F C60 (30 nm)	0.48	3.16	0.42	0.63

to the electron collecting electrode and the exciton to prevent the quench on the metal electrode. C60 has a deep highest occupied molecular orbital (HOMO) level at 6.5 eV. The hole blocking is provided by the energy barrier between HOMO of C60 and that of the polymers, MEH-PPV and P3HT, at 5.1 and 4.8 eV, respectively. These blocking of the C60 interlayer should bring the improvement in J_{sc} until the series resistance became too high by too thick layer at more than 10 nm.

4. CONCLUSIONS

We have investigated the polymer:PCBM bulk heterojunction photovoltaic cells with C60 interlayer. By inserting C60 interlayer (~10 nm), the both polymer cells with MEH-PPV:PCBM = 1:4 and P3HT:PCBM = 2:1 showed significant improvement in J_{sc} and η_p in approximately 10% compared to cells without the C60 layer.

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