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Improved photovoltaic characteristics of organic cells with heterointerface layer as a hole-extraction layer inserted between ITO anode and donor layer



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ABSTRACT

We have fabricated an improved organic photovoltaic (OPV) cell in which organic heterointerface layer is inserted between indium-tin-oxide (ITO) anode and copper-phthalocyanine (CuPc) donor layer in the conventional OPV cell of ITO/CuPc/fullerene (C₆₀)/ bathophenanthroline (Bphen)/Al to enhance the power conversion efficiency (PCE) and fill factor (FF). The inserted ITO-buffer layer consists of electron-transporting layer (ETL) and hole-transporting layer (HTL). We have changed the ETL and HTL materials variously and also changed their layer thickness variously. It is confirmed that ETL materials with higher LUMO level than the work function of ITO give low PCE and FF. All the double layer buffers give higher PCE than a single layer buffer of TAPC. The highest PCE of 1.67% and FF of 0.57% are obtained from an ITO buffer consisted of 3 nm thick ETL of hexadecafkluoro-copperphthalocyanine (F₁₆CuPc) and 3 nm thick HTL of 1,1-bis-(4-methyl-phenyl)-aminophenylcyclohexane (TAPC). This PCE is 1.64 times higher than PCE of the cell without ITO buffer and 2.98 times higher than PCE of the cell with single layer ITO buffer of TAPC. PCE is found to increase with increasing energy difference (ΔE) between the HOMO level of HTL and LUMO level of F_{16} CuPc in a range of $\Delta E < 0.6$ eV. From the ΔE dependence of PCE, it is suggested that electrons moved from ITO to the LUMO level of the electron-transporting F₁₆-CuPc are recombined, at the F₁₆CuPc/HTL-interface, with holes transported from CuPc to the HOMO level of HTL in the double layer ITO buffer ETL, leading to efficient extraction of holes photo-generated in CuPc donor layer.

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1. Introduction

Organic photovoltaic (OPV) cell has a basic structure where two layers of p-type and n-type conduction materials are sandwiched between two dissimilar electrodes [1]. The p-type layer works as a hole transport and electron

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donor layer, while n-type layer works as an electron transport and electron acceptor layer. This OPV cell is interested as solar energy harvest device because of advantages such as mechanical flexibility, easy fabrication, low fabrication cost, and light weight. Unlike inorganic solar cells, however, the power conversion efficiency (PCE) of OPV cells is low at this moment. Several efforts have been made to increase PCE. For example, cells with planar-heterojunction (PHJ), bulk-heterojunction (BHJ), and tandem layer structures have been studied [2–4].

Another effort has been made to improve the efficiency. It is insertion of buffer layer between anode (e.g., indiumtin-oxide (ITO)) and donor layer to avoid the energy level mismatch between work function of anode and the highest occupied molecular orbital (HOMO) energy of donor material. Additional buffer layer is inserted between cathode (e.g., Al) and acceptor layer to protect the acceptor layer from metal diffusion. Wide gap bathocuproine (BCP) and bathophenanthroline (Bphen) have been used as the cathode buffer because electrons from acceptor layer transport to cathode through defect states induced by thermalization of metal atoms of cathode [5–7].

Regarding the anode buffer layer attached to ITO, poly (ethyleneoxythiophene):poly(styrene-sulfonate) (PEDOT: PSS) has been widely used [8]. PEDOT:PSS, however, has disadvantages such as device degradation and electrical inhomogeneity because its acidity damages the ITO electrode [9-11]. Transition metal oxides like MoO₃, WO₃, V₂O₅, and NiO_x have been also used as ITO buffer [12,13,5,14]. Of these oxides, MoO₃ remarkably increases the work function of ITO, resulting in increase of PCE. In fact a high PCE was achieved BHJ cells [15-17]. The PHJ cell with ITO/MoO₃ connected to copper-phthalocyanine (CuPc) donor layer, however, does not give high efficiency [18]. Besides PEDOT:PSS and metal oxides, 1,1-bis-(4methyl-phenyl)-aminophenylcyclohexane (TAPC) hexadecafkluoro-copper-phthalocyanine (F₁₆CuPc) have been used as ITO buffer [19-22]. So far a single layer of TATC or F₁₆CuPc has been used as ITO buffer.

Adachi group obtained an electroluminescence (EL) performance using a heterointerface between F₁₆CuPc and 4,4′,4″-tris [3-methylphenyl (phenyl) amino triphenylamine] (m-MTDATA) in a hole-injection-free organic light emitting diode (OLED), which is higher than the cases of conventional double-injection OLED [23]. F₁₆CuPc works as electron-transporting layer (ETL) in this OLED, while m-MTDATA works as hole-transporting layer (HTL). Under impetus from this high performance, we study OPV cells with a new ITO buffer, i.e., double ITO buffer layer consisting of ETL and HTL, in the present work, investigation was mainly undertaken to answer the questions if such a double buffer layer gives rise to high PCE and what buffer material leads to high PV performance, and what mechanism works for the double ITO buffer.

OPV cells with a structure of ITO/CuPc/ C_{60} /Bphen/Al have been frequently studied [6,14,25]. CuPc and C_{60} (fullerene) are p- and n-type conduction materials (electron donor and acceptor), respectively, and Bphen is an electron-transporting cathode buffer which is also working as exciton-blocking layer due to wide HOMO–LUMO gap. Bphen rather than BCP was chosen because Bphen has a

higher electron mobility than BCP [26]. Here we investigate the effect of the heterointerface ITO buffer to the PCE of this cell. We insert a double layer of ETL/HTL between ITO and CuPc layer as ITO buffer, i.e., our OPV cell is ITO/ETL/HTL/CuPc/C₆₀/Bphen/Al. The ITO buffer of ETL/HTL works to extract holes from CuPc donor layer and to transport them to ITO. Therefore this buffer is called hole extraction layer (HEL). Two series of HEL are fabricated. The first series is that F_{16} CuPc is fixed as ETL material, while HTL material is changed variously. The second series HEL is that TAPC is fixed as HTL material, while ETL material is changed variously.

We found from study of the first series ITO buffers that (1) TAPC used as HTL gives a highest peak PCE of 1.67%, which is 1.64 and 2.98 times higher than the PCEs of the cells with bare ITO and single HEL of TAPC, respectively, and (2) PCE depends on the energy difference (ΔE) between HOMO of HTL ([HOMO]_{HTL}) and LUMO of F₁₆CuPc ([LUMO]_{F16CuPc}). We also found from study of the second series ITO buffers that the highest PCE is obtained when F₁₆CuPc is used as ETL.

2. Experimental methods

All organic thin films were grown on glass substrates precoated with a layer of ITO (sheet resistance: $10~\Omega/\text{sq}$) in a high vacuum chamber with base pressure of 5×10^{-4} Pa. The organic materials were obtained commercially and were used without further sublimation. Before the organic films were deposited, the substrates were treated by UV lamp for 15 min. Deposition rate of 1-2~Å/s was maintained for all organic materials, while 10~Å/s for Al cathode. The calibrated quartz crystal thickness monitor was used to measured thickness of each layer. The active area of the devices was about $0.3\times 0.3~\text{cm}^2$.

OPV cells with structure of ITO/ETL/HTL/CuPc (20 nm)/ C_{60} (40 nm)/Bphen (8 nm)/Al (100 nm) were constructed, where parenthesis indicates layer thickness. For the first series cell where ETL material was fixed to F_{16} CuPc, HTL material was changed among TCTA, 5,6,11,12-tetraphenylnaphthacene (rubrene), N,N-diphenyl-N,N-3-methylphenyl-1, 1-biphenyl-4,4-diamine (TPD), 3,4,9,10-perylenetetracarboxylicbisbenzimidazole (2-NTANA), m-MTDATA, 4,4-bis[N-(1-naphthyl)-N-phenyl-amino] biphenyl (NPB), pentacene, and 4,4,4-tris(N-carbazolyl-triphenyl amine (TCTA). For the second series cells where HTL material is fixed to TAPC, ETL material was changed among F_{16} CuPc, Bphen, tris(8-hydroxyquinoline)aluminum (Alq₃), 3,4,9,10-perylenetetracarboxylicbisbenzimidazole (PTCBI), and C_{60} .

Current–voltage $(J_{SC}-V_{OC})$ characteristics were measured with a programmable source meter (Keithley-2400) in the dark and under AM1.5 solar illuminations at intensity of $100 \, \text{mW/cm}^2$ of solar simulator. The light intensity was measured with a calibrated Si-solar cell. All measurements were carried out at room temperature under ambient conditions without any protective coating.

3. Results and discussion

Fig. 1a shows a schematic layer structure of our OPV cell and chemical structures of the HTL materials in the HEL

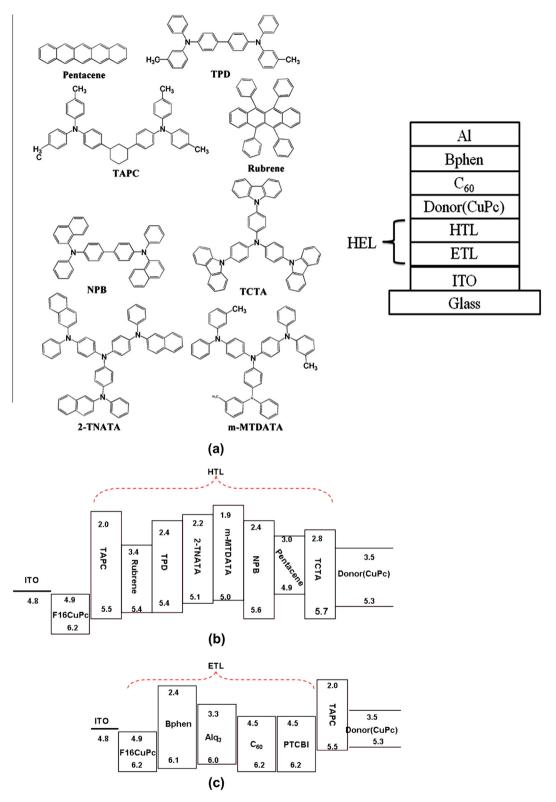


Fig. 1. (a) Schematic layer structure of our OPV cell and chemical structures of the HTL materials in HEL, (b) HOMO/LUMO energy levels of various hole-transporting materials used as HTL in the first series double-structure ITO buffer, and (c) HOMO/LUMO energy levels of various electron-transporting materials used as ETL in the second series ITO buffer.

Table 1PV parameters of the cells with the first series ITO buffer of hole extraction layer (HEL) where the ETL material is fixed to F_{16} CuPc but the HTL material is changed variously. ΔE is the energy difference between HOMO level of HTL and LUMO level of F_{16} CuPc.

Buffer layer of ITO	J _{SC} (mA/cm ²)	$V_{\rm OC}\left({\sf V}\right)$	FF (%)	PCE (%)	ΔΕ
ITO/TAPC	4.07	0.448	0.31	0.56	
Bare ITO	4.25	0.465	0.53	1.02	_
F ₁₆ CuPc /Pentacene	4.41	0.451	0.52	1.04	0
F ₁₆ CuPc/m-MTDATA	5.06	0.451	0.55	1.23	0.1
F ₁₆ CuPc/2-TNATA	5.33	0.413	0.55	1.27	0.2
F ₁₆ CuPc/TPD	5.89	0.463	0.53	1.42	0.5
F ₁₆ CuPc/Rubrene	5.59	0.516	0.54	1.48	0.5
F ₁₆ CuPc/TAPC	6.01	0.491	0.57	1.67	0.6
F ₁₆ CuPc/NPB	5.38	0.491	0.42	1.14	0.7
F ₁₆ CuPc/TCTA	4.33	0.443	0.47	0.91	0.8

with organic heterointerface layer. The LUMO and HOMO energy levels of the HEL materials used in the first and second series cells are shown in Fig. 1b and c, respectively.

Table 1 summarizes the PV parameters of OPV cells with the first series ITO buffers. The thickness of ETL (and also HTL) of HEL buffer in the cell was fixed to 3 nm. The PV performance of the cells without HEL (i.e., without ITO buffer, called bare ITO hereafter) and with a single buffer layer of TAPC (called ITO/TAPC) is also shown for comparison. A maximum PCE of 1.67% is obtained from HEL of F₁₆CuPc/TAPC, which is 1.64 and 2.98 times higher than the PCEs of the cells with bare ITO and single HEL of TAPC, respectively. From the present study we have realized that the double-layer ITO buffer gives higher PCE and FF than the single-layer ITO buffer.

Fig. 3a plots PCE against energy difference (ΔE) between [HOMO]_{HTL} and [LUMO]_{F16CuPc}. The PCE increases with increasing ΔE from 0 eV to 0.6 eV, while it decreases with increasing ΔE from 0.6 eV to 0.8 eV. The highest PCE of 1.67% is obtained at ΔE = 0.6 eV. It seems that the PCE increases nearly exponentially in a range of 0–0.6 eV.

The carrier hopping rate from donor to acceptor with their energy level separation of ΔE is described by the Miller–Abrahams rate model.

$$v_{ii} \propto v_0 \exp[-2\alpha r_{ii}] \exp[(\Delta E/K_B T)\theta(-\Delta E)]$$
 (1)

where v_0 is the attempt-to-escape frequency, α^{-1} is the localization radius of a charge carrier, K_B is the Boltz-

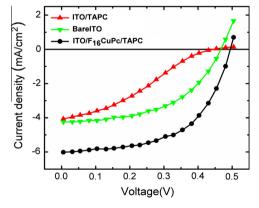


Fig. 2. Current–voltage $(J_{SC}-V_{OC})$ curves of the OPV cells with bare ITO (i.e., without HEL), ITO/TAPC (i.e., with a single HEL of TATC), and HEL of F_{16} CuPc/TAPC.

mann constant, T is the temperature and θ is the Heaviside function [24]. Taking into account that Heaviside function is a unit step function, it is expected that the hopping rate increases exponentially with increasing ΔE . Therefore it is suggested that the nearly exponential increase of PCE observed at $\Delta E < 0.6$ eV (Fig. 3a) is understood by the hopping process in the ITO buffer of HEL as follows.

Electrons from ITO move to the LUMO level of F₁₆CuPc layer in the ITO buffer. ITO is positively charged by the moving of electron. The electrons in F₁₆CuPc cannot move to the LUMO level of HTL in the ITO buffer because of high barrier but move to the HOMO level of HTL by hopping. The HOMO level is partially occupied by holes transported from CuPc layer. Therefore recombination of electrons from ITO and holes photo-generated in CuPc occurs at the heterointerface of F₁₆CuPc/HTL. This hole extraction process by electron-hole recombination is shown in Fig. 3b. Unlike the case of ΔE < 0.6 eV, the PCE decreases with increasing ΔE at $\Delta E > 0.6$ eV. The reason is not perfectly clear at this moment. As one of the reasons, moving of holes from HOMO level of HTL to the HOMO level of F16-CuPc might be conceivable. F₁₆CuPc is electron-transporting material but inefficient hole-transporting material, therefore the PCE is expected to become low. We will investigate the reason in detail in near future.

The current density (J_{SC})–voltage (V_{OC}) characteristics of cells with bare ITO, ITO/TAPC, and HEL of F_{16} CuPc/TAPC are shown in Fig. 2. From this figure, we understand that PCE increases from 0.56% to 1.02% and to 1.67% when the ITO buffer changed from ITO/TAPC to bare ITO and to HEL of F_{16} CuPc/TAPC. An S-shape J_{SC} – V_{OC} curve is observed in the case of ITO/TAPC. It is suggested that this shape is due to energy level mismatch between TAPC and ITO anode (see Fig. 1b). Such a mismatch leads to a considerable decrease of both the fill factor (FF) and PCE [23]. In fact the cell with ITO/TAPC shows lower FF of 0.31% and smaller PCE of 0.56% than the cells with double HEL (Table 1).

Table 2 summarizes the PV parameters of OPV cells with the second series ITO buffers where HTL material of HEL is fixed to TAPC but ETL material is changed variously. All the ETL materials (Bphen, Alq $_3$, C $_6$ 0, and PTCBI) give much lower PCE than F $_16$ CuPc. Unlike F $_16$ CuPc, the LUMO levels of Bphen, Alq $_3$, C $_6$ 0, and PTCBI are higher than the work function of the ITO (Fig. 1c). Therefore the electrons from ITO are difficult to reach ETLs/TAPC-interface, leading to less recombination possibility.

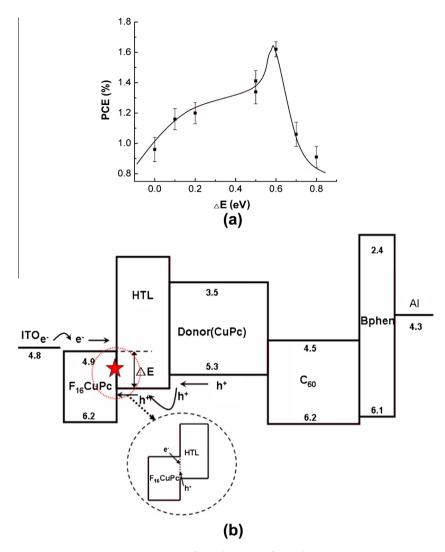


Fig. 3. (a) PCE plotted against the energy separation (ΔE) between [LUMO]_{F16CuPc} and [HOMO]_{HTL}. (b) A schematic diagram showing the recombination process of holes from the donor CuPc and electrons from the ITO anode in the OPV cell used in the present work. All level energies are expressed in electron volts (eV).

Table 2PV parameters of the cells with the second series ITO buffer of HEL where the HTL material is fixed to TAPC but the ETL material is changed variously.

ETL	J _{SC} (mA/cm ²)	V _{OC} (V)	FF	PCE (%)
Bphen	5.26	0.44	0.47	1.1
Alq_3	4.94	0.43	0.48	1.06
C ₆₀	4.19	0.41	0.48	0.83
PTCBI	5.07	0.35	0.51	0.85

We changed the layer thickness of $F_{16}\text{CuPc}$ variously in the OPV cell with HEL of $F_{16}\text{CuPc}/\text{TAPC}$ but keeping the same thickness for $F_{16}\text{CuPc}$ and TAPC. Its PV characteristics are summarized in Table 3. The PCE increases with increasing the layer thickness from 0.5 nm to 3.0 nm, while it decreases with increasing the thickness from 3.0 nm to 10.0 nm. The highest PCE is obtained from the cell with 3 nm thick $F_{16}\text{CuPc}$ layer and 3 nm thick TAPC layer. This result is understood as follows. In case of thin ETL and

Table 3 PV parameters of the cells with ITO heterointerface buffers with various $F_{16}\text{CuPc}$ and TAPC layer thicknesses.

F ₁₆ CuPc(xnm)	TAPC(ynm)	J _{SC} (mA/cm ²)	$V_{\rm OC}\left({\sf V}\right)$	FF	PCE (%)
0.5	0.5	4.56	0.473	0.51	1.12
1.0	1.0	4.86	0.485	0.54	1.19
2.0	2.0	5.73	0.491	0.57	1.58
3.0	3.0	6.01	0.491	0.57	1.67
4.0	4.0	4.98	0.482	0.57	1.41
7.0	7.0	4.82	0.483	0.47	1.07
10.0	10.0	4.33	0.491	0.46	1.03

HTL layers, formation of consecutive films is so difficult that a complete ETL/HTL-interface is not formed, while in case of thick layers the series resistance of the cells increases, resulting in decrease of PCE and FF.

Table 4 summarizes the hole mobilities of various holetransporting materials used in the cells with the second series ITO buffers. It is found that the mobility does not

Table 4Hole mobilities of various hole-transporting materials, together with their HOMO level energies.

	CuPc	TAPC	Rubrene	TPD	2-NTATA
Hole mobility (cm ² V ⁻¹ s ⁻¹) HOMO (eV)	$\begin{array}{c} 2.4\times 10^{-4}[27] \\ 5.3[27] \end{array}$	$\begin{array}{c} 2.45 \times 10^{-3} [28] \\ 5.5 [28] \end{array}$	$1.0 \times 10^{-6} [29] \\ 5.4[29]$	$1.4 \times 10^{-3} [28]$ $5.4[28]$	$6.40\times 10^{-5} [28] \\ 5.1[28]$
	m-MTDATA	NPB		Pentacene	TCTA
Hole mobility (cm 2 V $^{-1}$ s $^{-1}$) HOMO (eV)	3 × 10 ⁻⁵ [30] 5.0[31]	2.2 × 10 5.6[32]		$0.35 \times 10^{-2} [27] \\ 4.9[27]$	3 × 10 ⁻⁴ [33] 5.7[33]

give influence to the PCE. For example, (1) pentacene shows lower PCE of 1.04% than the other materials although it has the highest mobility of the various materials, and (2) rubrene shows higher PCE of 1.48% than the other materials except TATC although it has the lowest mobility. Therefore it is suggested that ΔE between the energies of [HOMO]_{HTLs} and [LUMO]_{F16CuPc} is important factor in improving PCE.

4. Conclusions

The PV characteristics are studied for OPV cells of ITO/CuPc/ C_{60} /Bphen/Al with double layer HEL as ITO buffer, which consists of ETL and ETL, by changing the ETL and HTL materials variously and changing the layer thickness variously. The ITO buffers with such a heterointerface layer structure show higher PCE and FF than the ITO buffer with a single layer. A highest PCE of 1.67% and FF of 0.57% are obtained from ITO buffer consisted of 3 nm thick electron-transporting F_{16} CuPc and 3 nm thick hole-transporting TAPC. This PCE is 1.64 and 2.98 times higher than the PCEs of the cells with bare ITO and single HEL of TAPC, respectively.

It is found that PCE increases with increasing energy difference ΔE between the HOMO level of HTL and LUMO level of F₁₆CuPc in HEL in a range of $\Delta E < 0.6$ eV. From the observed ΔE dependence of PCE, we have proposed that electrons moved from ITO to the LUMO level of the electron-transporting F₁₆CuPc are recombined, at the F₁₆-CuPc/HTL-interface, with holes transported from CuPc to the HOMO level of HTL in the double layer ITO buffer ETL. It is experimentally confirmed that ETL materials with higher LUMO level than the work function of ITO give lower PCE and FF than F₁₆CuPc with almost the same LUMO level as the work function of ITO.

Our findings provide an important information to enhance the extraction of holes generated in donor CuPc and acceptor C_{60} layers and to achieve a high PCE of OPV cell.

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