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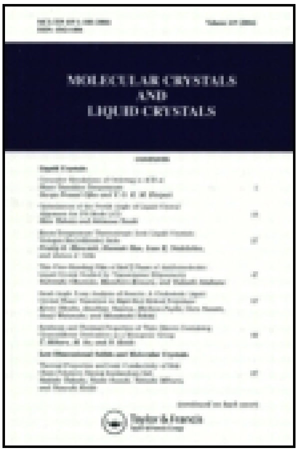

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High Mobility Hole Extraction Material for Organic Solar Cell Application

Gyeong Woo Kim [a], Woo Sik Jeon [a], Young Hoon Son [b], Jang Hyuk Kwon [a] & Sung-Hyun Jung [b]

[a] Department of Information Display, Kyung Hee University, Dongdaemoon-gu, Seoul, 130-701, Republic of Korea

[b] Cheil Industries Inc., Gocheon-dong, Uiwang-si, Gyeonggi-do, 332-2, Republic of Korea

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High Mobility Hole Extraction Material for Organic Solar Cell Application

GYEONG WOO KIM,[1] WOO SIK JEON,[1] YOUNG HOON SON,[2] JANG HYUK KWON,[1,*] AND SUNG-HYUN JUNG[2]

[1]Department of Information Display, Kyung Hee University, Dongdaemoon-gu, Seoul 130–701, Republic of Korea
[2]Cheil Industries Inc., Gocheon-dong, Uiwang-si, Gyeonggi-do, 332–2, Republic of Korea

We report a high mobility hole extraction material, N,N,N′*,N*′*,-tetrakis(biphenyl-4-yl)benzidine (TBBD) for organic solar cell applications. Its highest occupied molecular orbital energy value at 5.3 eV is well matched with a donor layer and its high mobility of 1.74* × *10^{-2} cm^2/Vs brings out an easy and rapid extraction of holes from the donor material. The TBBD incorporated device showed significant improvement of open-circuit voltage, resulting in power conversion efficiency improvement of 9% compared with reference device.*

Keywords Organic solar cells; Hole extraction layer; Power conversion efficiency; Mobility; Built-in potential

Introduction

The performances of organic solar cells (OSCs) have advanced rapidly in recent years due to development of new materials which has shown their ability to harvest solar energy effectively [1,2]. Substantial efforts to improve the key factors such as power conversion efficiency (PCE) of OSCs have been made in recent days with growing demand for the low cost power sources [3,4]. Recent reports show that the PCE is dependent on both photocurrent and photovoltage [5–8]. The open-circuit voltage (V_{oc}), one of the key parameters, is still a debatable issue in the OSCs. Until now, the models of V_{oc} based on the energy difference between the highest occupied molecular orbital (HOMO) or ionization potential of the donor and the lowest unoccupied molecular orbital (LUMO) or electron affinity of the acceptor materials [9], the choice of electrode materials [10], work function difference between the two electrodes [11–13], active layer thickness [14], light intensity and temperature dependent generation of free charge carriers have been considered [15]. Furthermore efficient charge extraction to improve the charge transport between the organic layer and electrode is indispensable, which can avoid the loss of energy level offset. Recent advances have shown that hole extraction layers (HELs) as a buffer layer are decisive in determining the enhanced V_{oc} in OSCs [16–19] by minimizing this energy losses. High charge carrier

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[*] Address correspondence to Prof. Jang Hyuk Kwon, Department of Information Display, Kyung Hee University, Dongdaemoon-gu, Seoul 130–701, Republic of Korea. Tel.: (+82)-2–961-0948; Fax: (+82)-2–968-6924. E-mail: jhkwon@khu.ac.kr

mobility and proper HOMO level of HEL are the key parameters for controlling the V_{oc} loss leading to a point of enhancing efficiency of OSCs. Nonetheless, very few HEL materials with compatible HOMO level and high mobility have been synthesized so far [19]. In this paper we present a new HEL material, TBBD for OSCs application. The TBBD as a buffer layer influences the V_{oc} and PCE directly due to its high mobility and favorable HOMO level with subphthalocyanine chloride (SubPc) donor for efficient charge extraction.

Experimental

Synthesis

N, N-Di-(4-biphenylyl)benzylamine (3). To a solution of 4-bromobiphenyl (10 g, 42.9 mmol), sodium t-butoxide (4.32 g, 44.9 mmol), palladium acetate (42 mg, 0.187 mmol), benzylamine (2.04 ml, 18.7 mmol) and to tris-t-butylphosphine (469 μl, 0.374 mmol) were added to 60 mL of dehydrated toluene. After stirred at 120°C for 7.5 hours, the reaction mixture was allowed to cool to room temperature for one night under the atmosphere of argon. 300 ml of dichloromethane was added and precipitates were dissolved. After washing with 60 ml of a saturated aqueous solution of sodium chloride, the organic layer was dried with anhydrous potassium carbonate. After potassium carbonate was removed by the filtration, the organic solvent was removed by distillation. To the obtained residue, 200 ml of toluene and 40 ml of ethanol were added and the residue was completely dissolved by heating at 80°C. The resultant solution was slowly cooled to the room temperature by being left standing for one night and the recrystallization was conducted. The formed crystals were separated by filtration. After drying the crystals, 6.73 g (16.4 mmol) of N,N-di(4-biphenylyl)benzylamine was obtained at a yield of 87%.

Di-4-biphenylylbenzylamine (4). To a solution of N,N-di(4-biphenyl)benzylamine (1.35 g, 3.28 mmol), palladium-active carbon (135.00 mg, 10% by weight) was added to 100 mL of chloroform and 20 mL of ethanol. And then, 2 liters of hydrogen gas purged to solution. After the fluid was stirred for the total time of 30 hours at the room temperature, 100 ml of dichloromethane was added and catalyst was removed by filtration. The obtained solution was washed with 50 ml of a saturated solution of sodium hydrogen carbonate. The organic layer was separated and dried with anhydrous potassium carbonate. After filtering potassium carbonate, the solvent was removed by distillation. To the obtained residue, 50 ml of toluene was added and the recrystallization was conducted. The formed crystals were separated by filtration and dried in vacuum at 50°C. And then, 990 mg (3.08 mmol) of di-4-biphenylylamine was obtained at a yield of 94%.

TBBD (5). To a solution of di-4-biphenylylamine (4.33 g, 13.46 mmol), 4,4′-dibromobiphenyl (2.00 g, 6.41 mmol), bis(dibenzylideneacetone)palladium(0) (0.22 g, 0.38 mmol), sodium t-butoxide (7.76 g, 80.77 mmol) and tris-t-butylphosphine (0.08 g, 0.38 mmol) were added to 100 mL of dehydrated toluene under nitrogen. After stirred at 115°C for 12 hours, the reaction mixture was allowed to cool to room temperature for overnight. The formed precipitates were completely dissolved in 4 L of dichloromethane. After it was washed with 1 L of a saturated aqueous solution of sodium chloride, the organic layer was dried with anhydrous potassium carbonate. And then, the organic solvent was removed by distillation and 1.3 L of toluene and 400 ml of ethanol was added to the obtained residue. The precipitates were dissolved by heating at 80°C and the resultant mixture was slowly cooled to the room temperature. The formed crystals were washed with

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small amounts of toluene and ethanol and dried at 60°C for overnight in a vacuum drying oven. And then, 3.7 g of TBBD was obtained at a yield of 67.6%. ^{1}H NMR δ_H(400 MHz; $CDCl_3$) 7.56 (8H, d), 7.52 (12H, d), 7.43 (8H, t), 7.32 (4H, t), 7.24 (12H, d). IR (KBr; cm^{-1}) 3031 (aromatic C-H), 1277, 1322 (aromatic C-N), 1597 (aromatic C = C), M/S (m/z, M$^+$): Found: 792.72, Calc. For $C_{60}H_{44}N_2$: 792.35.

Fabrication

OSCs were fabricated on a clean glass substrate precoated with a ITO layer which has 150nm thickness with a sheet resistance 15 Ω/square was used. The patterns of 2 × 2 mm^2 active area were formed by photolithography processes. Before organic layer deposition process, the glass substrate was cleaned by sonification in an acetone, isopropyl alcohol, and deionized water for 10 minute respectively. After drying it by blowing nitrogen gas, it was finally irradiated in a UV-ozone chamber. The ozone gas was generated by UV light to excite the oxygen in the air inside the chamber. The organic materials TBBD, SubPc (Lumtech), fullerene (C_{60}) (Alfa Aesar), and bathocuproine (BCP) (Lumtech) were sequentially deposited onto ITO with a deposition rate of 0.3Å/s under a pressure of $\sim 3.0 \times 10^{-7}$ Torr. Subsequently, Al (iTASCO) was deposited with a deposition rate of ~2.5 Å/s under a pressure of $\sim 1 \times 10^{-6}$ Torr.

Measurements

The current density–voltage (J–V) characteristics of the fabricated devices were measured using a computer controlled Keithley 2400 source-meter, in the dark and under illumination intensity of 100 mW/cm^2 with calibrated AM 1.5G sun simulator (Asahi Japan, Model HAL-302) in ambient conditions. A xenon light source was used to give simulated irradiance of 100mW/cm^2 (equivalent to an AM1.5G irradiation) at the surface of the device. The J-V characteristic measurements under illumination were carried out in a dark chamber with a window slit of 5 mm^2 area for illumination. UV-vis absorption spectrum of TBBD molecule was recorded with a Shimadzu 3100 UV-vis-NIR spectrophotometer. Cyclic voltammetry was performed using EC epsilon electrochemical analysis equipment. Platinum wire and ITO film on glass were used as a counter and working electrodes, respectively, while tetrabutylammonium perchlorate (Bu_4NClO_4) was used as the supporting electrolyte. Also, Ag wire in 0.1M $AgNO_3$ was used as a reference electrode. Using an internal ferrocenium/ferrocene(Fe$^+$/Fe) standard, the potential values were converted to the saturated calomel electrode(SCE) scale. The molecular structure was confirmed by ^{1}H NMR (BrukerAvance 400 NMR spectrometer), mass spectrometry and FT-IR (Varian FTS 700).

Results and Discussion

The TBBD molecule (**5**) was synthesized according to the Scheme 1. The initial product of *N,N*-di-(4-biphenyl)benzylamine (**3**) was synthesized by the reaction of benzylamine (**1**) and 4-bromobiphenyl (**2**) as the starting materials. Subsequent treatment of (**3**) with hydrogen in palladium active carbon in chloroform and ethanol mixed solvents gives the crystal of di-4-biphenylyl-amine (**4**) in a yield of 94%. Reaction of (**4**) with 4,4′-dibromobiphenyl gives the TBBD (**5**) as the final product. To identify HOMO and LUMO levels of the synthesized TBBD, cyclic voltammetry and absorption spectrum were investigated. From the onset of oxidation potential of cyclic voltammetry with comparison of the HOMO

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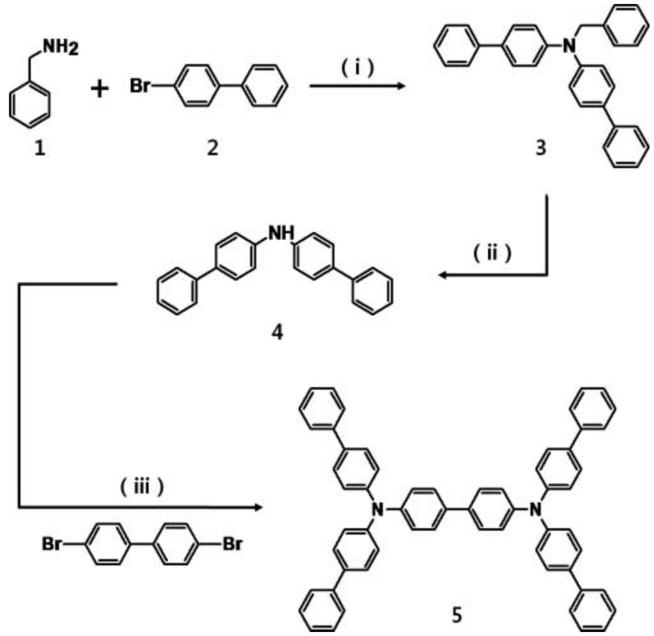

Scheme 1. Synthesis of TBBD molecule. Reagents and conditions: (i) $Pd(OAc)_2/P(tBu)3$, NaOtBu, 120°C, 7.5 hr; (ii) H_2, 10% Pd/C, $CHCl_3/EtOH$ (5:1), room temp.,30 hr; (iii) $P(tBu)_3$, $Pd(dab)_2$, NaOtBu, 115°C, 12 hr.

energy level of ferrocene (4.8 eV), TBBD's HOMO energy level was found to be at 5.3 eV. The optical band gap was calculated as 3.1 eV from the absorption edge of absorption spectrum. Finally, the LUMO energy level was determined to be 2.2 eV by the calculation from a HOMO value. The glass-transition temperature (T_g) and melting temperature (T_m) of the TBBD material were found to be at 135.5°C and 267°C by employing the thermal gravimetric analysis (TGA), and differential scanning calorimetry (DSC).

Figure 1 shows the absorption and emission spectra of the TBBD in chloroform. The absorption spectrum of TBBD gives a broad absorption band peak at 357 nm, which could

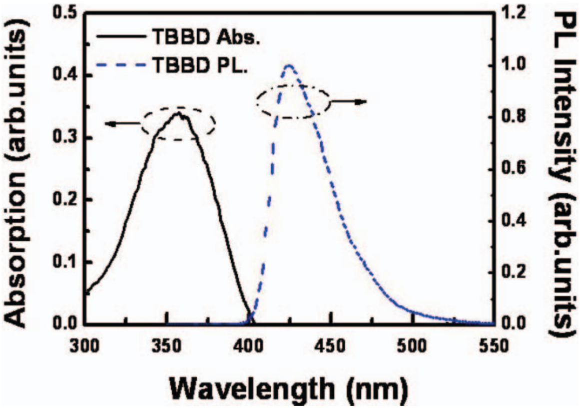

Figure 1. Absorption and photoluminescence (PL) spectra of TBBD molecule in chloroform solution.

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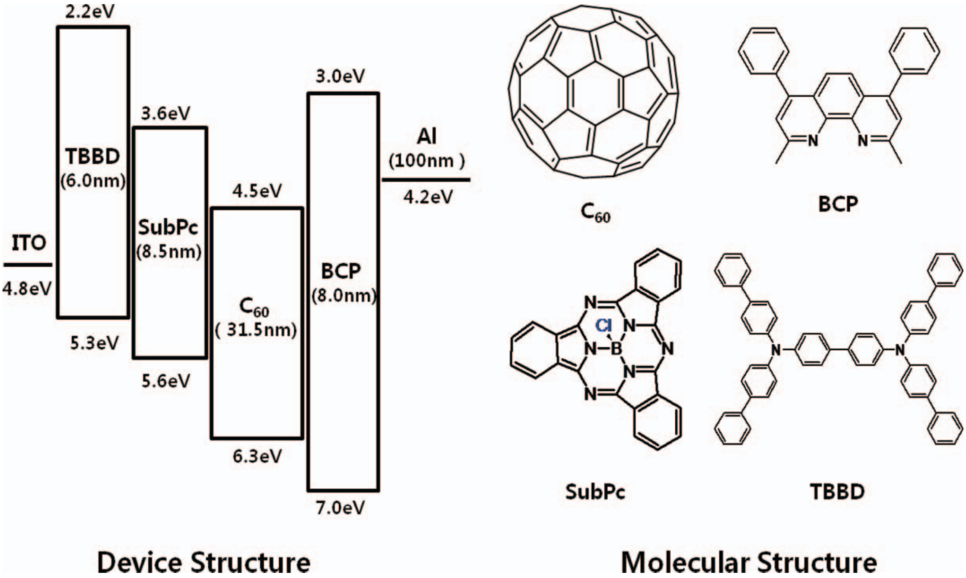

Figure 2. Device architecture and molecular structure of fabricated OSCs.

be π-π^* transitions of the conjugated π-electron system. The TBBD material exhibits a photoluminescence emission (PL) at 425 nm. The TBBD is slightly soluble in chloroform and is insoluble in almost all organic solvents.

Figure 2 shows the device and molecular structures of fabricated OSCs. To investigate the J-V characteristics with and without TBBD HEL in planer hetero-junction SubPc/C60 OSCs, the devices were optimized with varying the donor and HEL thickness. Optimized OSCs with and without TBBD HEL layer were as follows:

Reference device : ITO/SubPc(14.5nm)/C_{60}(31.5nm)/BCP(8.0nm)/Al(100nm)
HEL device : ITO/HEL(6.0nm)/SubPc(8.5nm)/C_{60}(31.5nm)/BCP(8.0nm)/Al(100nm).

The J-V characteristics with and without TBBD are shown in Figure 3. The SubPc/C_{60} reference device shows the V_{oc} of 0.89V, fill factor (FF) = 58.47, short-circuit current (J_{sc}) = 5.22 mA/cm^2, and PCE = 2.70%. The J-V characteristics of reference device have a terrible S-kink because of imbalanced carrier mobility between donor and acceptor. Low mobility of SubPc($\sim 10^{-5}$ cm^2/Vs) and high electron mobility of C_{60}($\sim 10^{-3}$ cm^2/Vs) makes the asymmetric field distribution which result in a terrible S-kink.[20, 21, 22] Our reference device was optimized with a terrible S-kink. For the high efficiency of reference device, S-kink could move after V_{oc}. On the other hand, J-V characteristics of HEL device show much relieved S-kink. In our HEL device, there still remains S-kink around V_{oc} region. The insertion of TBBD HEL layer significantly enhances the V_{oc} value to 1.16 V. However, insertion of HEL layer gives a lower FF as 46.96% compared to that of reference device. The J_{sc} and PCE were improved to 5.39 mA/cm^2 and 2.94%, respectively. The TBBD's high mobility seems to be very useful to attain very high J_{sc} with high PCE due to its efficient charge collection at the electrode. Furthermore a large portion of the benefit is gained by enhancing the V_{oc}. Table 1 lists the OSC characteristics of our devices.

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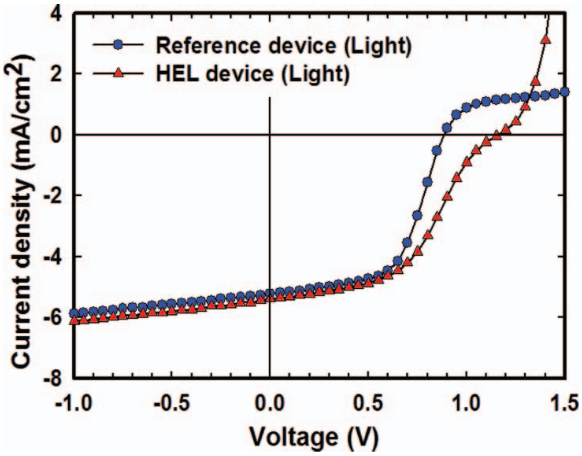

Figure 3. Current-Voltage characteristics of OSCs with and without TBBD layer under illumination condition.

In spite of the amount of light absorption by the donor layer in HEL device is decreased due to the reduced thickness, J_{sc} value is higher than reference device. It implies that introduction of TBBD enhances the performance of carrier transportation. This result is well matched the reduction of series resistance as shown in Table 1. The series resistance and shunt resistance of reference device and HEL device were measured by the J-V characteristics under dark condition [23]. To understand mobility effect and lower series resistance by the TBBD layer, hole mobility was measured by using space charge limited current (SCLC) [24]. Fabricated hole only device has ITO/MoO_3(0.75nm)/HEL(100nm)/MoO_3(5nm)/Al structure. The thin MoO_3 layer on metal electrode was inserted to make ohmic contact so that SCLC region can be investigated clearly. Mobility of TBBD was compared with NPB (4,4′-bis[*N*-(1-naphthyl)-*N*-phenyl-amino]biphenyl) material to see the difference. The SCLC carrier mobility depending on electric field was plotted, as shown in the Figure 4. The TBBD SCLC hole mobility is evaluated as $\sim 1.74 \times 10^{-2}$ cm^2/Vs at the 0.3 MV/cm^2 electric field. It is about two order magnitudes greater than that of a reference NPB ($\sim 2.18 \times 10^{-4}$ cm^2/Vs). Our calculated NPB mobility is very similar to the previously reported value [25].

Interesting V_{oc} increased result with the introduction of TBBD HEL in the OSC device was observed. The V_{oc} increase by increased J_{sc} in our HEL inserted devices could

Table 1. Summary of device performance

	Reference device	HEL device
Open-circuit voltage, V_{oc} (V)	0.89	1.16
Short-circuit current, J_{sc} (mA/cni^2)	58.47	46.96
Fill factor, FF (%)	5.22	5.39
Power conversion efficiency, PCE (%)	2.70	2.94
Series resistance, R_s ($_{dark}$) (Ω cm^2)	308.2	14.7
Shunt resistance, $R_{sh\,(dark)}$ (Ω cm^2)	5.5×10^5	3.1×10^6

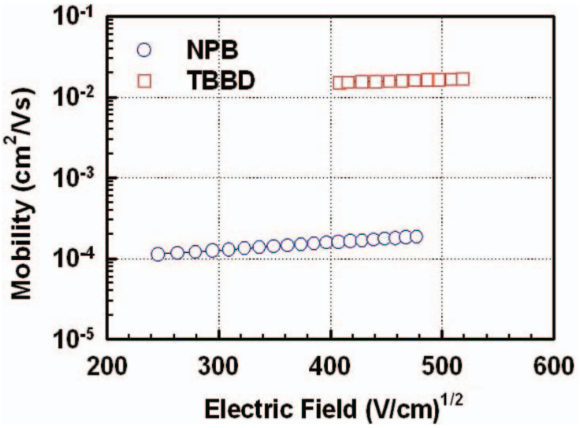

Figure 4. Hole mobility characteristics of TBBD and NPB layers.

understand from the diode equation and built-in potential voltage (V_{bi}). Reported diode equation [26] is as follows:

$$V_{oc} = \frac{nKT}{q} \ln\left(\frac{Jsc}{Jso}\right) + \frac{\Delta E_{DA}}{2q} \tag{1}$$

Where, J_{SO} is the reverse saturation dark current, *n* is ideality factor, ΔE_{DA} is the thermal activation energy for charge separation at the donor-acceptor interface. In addition to this equation, the V_{oc} depends linearly on the work function difference $\Delta\Phi_{electrodes}$ between the two electrodes. This V_{bi} is the voltage required for flat band condition which plays a key role in diffusing the charge carriers to the electrodes. High mobility HTLs can extract holes easily from donors layer because of better hole carrier mobilities than SubPC ($\sim 2\times10^{-5}$ cm^2/ Vs), which results in increase of J_{sc} with V_{oc}. On the other hand, the V_{oc} increase by increased V_{bi} in our OSCs could be explained by the following reasons. Correct positioning of the HOMO level with respect to donor material and efficient hole extraction at the SubPc/HEL heterojunction increases the concentration of charge densities at the interface, causing the redistribution of electric field within the device leading to the V_{bi} increase. To support our explanation about V_{oc} increase, on-set voltages between reference and HEL devices are compared from dark J-V characteristics. An on-set voltage of HEL device is about 0.95 V, while a reference device has $\sim$0.8 V. These results support that V_{bi} is increased by TBBD layer in our OSCs. The highest V_{oc} of 1.16 V achieved in HEL device is equivalent to a difference of the HOMO level of SubPc donor and LUMO level of C_{60} acceptor. This value corresponds to the maximum possible increase of V_{bi} inside. For this increase of V_{bi}, a little high HOMO level compared to donor material must be good because the energetically extraction force for the hole carrier movement should be considered. The high mobility is also exceedingly important for getting high J_{sc} and PCE. The TBBD's high mobility is very useful to attain high J_{sc} level due to its efficient charge collection at the electrodes.

Conclusion

In conclusion, we have presented a new hole extraction material, TBBD, for flat heterojunction OSCs. The mobility value of TBBD molecule was found to be 1.74×10^{-2} cm^2/Vs,

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which is very high to extract the holes smoothly from the donor materials in OSC devices. The TBBD incorporated device exhibits a power conversion efficiency of 2.94% along with a very high V_{oc} of 1.16V under 100mV/cm^2 simulated sunlight. The TBBD will be a very important hole extraction material to be implemented with other OSC applications.

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