

Evaluation of Small Molecules as Front Cell Donor Materials for High-Efficiency Tandem Solar Cells

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Organic solar cells (OSCs) with single-junction structures have made great progress in the past decade and power conversion efficiencies (PCEs) over 10% have been achieved through material innovation and device optimization.^[1-8] Despite the rapid advancement, single-junction OSCs suffer mainly from two losses: (i) the transmission of unabsorbed photons, which is due to the transmission of photons through the cell having a relatively lower energy than the bandgap energy, and (ii) the thermalization of excited carriers, which occurs when photons with a higher energy than the energy bandgap of the organic semiconductor are absorbed.^[9] A feasible and successful method to overcome such losses and further improve the PCE is to use tandem cells in which two or more cells with complementary absorption spectra are stacked on top of each other and connected in series.^[10,11] In recent years, great strides have been made on tandem cells and PCEs of 11.3%^[12] and 11.83%^[13] have been achieved for the double junction and three junction tandem cells, respectively. Much efforts in developing high efficiency tandem cell have focused on two aspects. One is to design active layer materials with complementary absorption.^[14-17] The other is to develop effective interconnection layers (ICLs), which could efficiently collect holes from one subcell and electrons from the other subcell, and then the two types of charge carriers recombine in the ICLs. Indeed, many efficient ICLs have been developed and successfully used in tandem cells.^[12,18-25] Nevertheless, the main technical obstacle in achieving higher performance with tandem OSCs is the active layer material. For serial connected tandem OSCs, making the open-circuit voltages (V_{oc}) as high as possible for

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each subcell is one requirement, since ideally the overall V_{oc} of the tandem cell equals the sum of $V_{\rm oc}$ of the sub-cells if there are no potential losses in the ICLs. Besides, the short-circuit current density (I_{sc}) of a tandem device is dictated by the lower value of the constituent subcells.^[26,27] Consequently, ideal tandem devices should have $V_{\rm oc}$ and $J_{\rm sc}$ as high as possible for each subcell.

In the past years, more attention has been focused on the design of low bandgap polymers with excellent absorption in the long wavelength range for the rear subcells.^[28-32] However, wide bandgap materials with both high V_{oc} and J_{sc} for the front cells are rare, which is becoming a serious limiting factor for the overall performance of tandem devices. For example, poly(3-hexylthiophene) (P3HT) has been widely used as the front cell donor of tandem OSCs, but generally with a low $V_{\rm oc}$ of ≈0.6 V when the most popular phenyl-C61-butyric acid methyl ester (PC₆₁BM) was used as the acceptor. Even when indene- C_{60} -bisadduct (ICBA) was used as the acceptor, the V_{oc} was only marginally improved to 0.84 V.[14,19,20] And more importantly, the single-junction cell based on P3HT for both cases still delivered a relatively low J_{sc} of around 10 mA cm⁻², which limits the overall J_{sc} and, therefore, the PCE of tandem OSCs.^[33] As has been widely accepted, small molecules (SMs) based OSCs have the advantages of well-defined structures, no batch-to-batch variation, versatile chemical structures which enables more facile control over the energy levels and generally higher V_{oc} , and so on.^[34,35] However, studies on tandem cells using SMs are much limited compared with that using polymer materials, and most tandem cells based on SMs are using the vacuum evaporation process.^[36-38] It would be advantageous if a simple and costeffective solution process could be used for SM-based tandem cells.

Recently, we have developed a series of oligomer-like SM donors for single-junction OSCs and the devices based on those SMs showed good photovoltaic performances with high Voc (≈0.9 V).^[39–41] In addition, most of the SMs show absorption in the range of 400-700 nm and devices based on these SMs showed J_{sc} s over 13 mA cm⁻² and external quantum efficiencies (EQE) values over 70%.^[39–41] These properties prompted us to investigate those SMs as donor materials in front subcells of tandem cells.

In this study, three representative SMs including DR3TSBDT, DR3TBDTT, and DRBDT-TT (Figure 1a) with high performance in single-junction structures were selected as the front cell donor materials in tandem cells. To have the best match for these front cell materials, one of the best-performed and repeatable polymer poly[4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b:4,5-b']dithiophene-co-3-fluorothieno-[3,4-b]thiophene-2-carboxylate], commonly known as PTB7-Th^[1]

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Figure 1. a) Molecular structures of SMs (DR3TSBDT, DR3TBDTT, and DRBDT-TT). b) Molecular structure of PTB7-Th. c) Normalized UV–Vis absorption spectra of SMs and PTB7-Th. d) Device structure of the tandem solar cell.

(Figure 1b), was selected as the rear cell donor, since the device based on this polymer showed both a high V_{oc} and high EQE values in the range 400–800 nm, particularly in the long-wavelength range (see Figure 1c). Phenyl-C71-butyric acid methyl ester (PC₇₁BM) was used as the acceptor for all subcells in this study. Consequently, these three SMs provided a high V_{oc} of over 1.65 V and J_{sc} of over 10 mA cm⁻² in all the tandem OSCs. The tandem solar cells based on DR3TSBDT:PC₇₁BM achieved a high PCE of 11.47%, and tandem solar cells using DR3TBDTT:PC₇₁BM and DRBDT-TT:PC₇₁BM as the front cells had PCEs of 10.73% and 10.43%, respectively. These results demonstrated that the oligomer-like SMs could offer a great choice as the front cell donor materials for tandem solar cells.

The absorption spectra of these SMs and PTB7-Th are shown in Figure 1c. These SMs show similar absorption within the range of 300-700 nm, and the PTB7-Th film shows strong absorption from 650 to 800 nm with a maximum at 710 nm, which complements the absent absorption in this range for the SM donors in the front subcell. The device structure of the tandem solar cell is shown in Figure 1d. Copper(I) thiocyanate (CuSCN)^[42] was used to modify the interface of ITO and the active layer due to its high optical transparency and good hole-transporting properties. ETL-1 (its structure shown in Figure S1, Supporting Information) was used to modify the interface of active layer and Al, a methanol-soluble fullerenesurfactant developed by Alex K.-Y as an efficient interfacial layer for cathodes.^[43] After a series of tests (detailed in Table S1, Supporting Information), the subcells were found to be best connected via an ICL of a 30 nm layer of ZnO nanoparticles processed from n-butanol^[44] and a 20 nm layer of pH neutral PEDOT:PSS (n-PEDOT:PSS) spin coated from aqueous solution (PEDOT: poly(3,4-ethylenedioxythiophene); PSS: polystyrene sulfonate). Importantly, using this optimized process, none of the solution processing steps caused damage to the

layers underneath, which is necessary to achieve high performance tandem solar cells. $^{\rm [45]}$

First, a thorough investigation of DR3TSBDT-based tandem solar cells was carried out. The subcell based on DR3TSBDT was fabricated using the similar procedures reported before,^[39] and the active layer based on PTB7-Th:PC71BM was processed using the optimized spin-coating with a mixed solvent of chloroform/1,8-diiodoctane (98.5%:1.5% by volume) solution (with a total donor and acceptor concentration of 12.5 mg mL⁻¹ and donor/acceptor weight ratio of 1:1.5) at 500 rpm 1.5 s and 1700 rpm for 20 s. The EQE spectra of DR3TSBDT:PC71BM and PTB7-Th:PC71BM single-junction devices are shown in Figure S3 (Supporting Information). All devices showed high EQEs in excess of 70%. Through systematic investigation, the optimized tandem cell was fabricated with an active layer thickness of 90 nm for the DR3TSBDT:PC71BM subcell, and this is not the same as that in the optimized single-junction solar cell, which showed a PCE of 9.95%, with V_{oc} of 0.92 V, a J_{sc} of 14.61 mA cm⁻² and FF of 0.74.^[39] The typical *J*–V characteristics of the optimized tandem solar cell and corresponding singlejunction reference subcells are shown in Figure 2 and Table S2 (Supporting Information), and the performance parameters are summarized in Table 1. With these conditions, the corresponding single-junction cell based on DR3TSBDT:PC71BM shows a PCE of 8.93% with $V_{oc} = 0.915$ V, $J_{sc} = 13.19$ mA cm⁻², and fill factor (FF) = 74.1%. And the single-junction cell based on PTB7-Th:PC₇₁BM delivers a PCE of 8.75% with V_{oc} = 0.80 V, $J_{sc} = 16.08$ mA cm⁻² and FF = 68.0%. Using these conditions, the optimized tandem solar cell with the structure of ITO/CuSCN/DR3TSBDT:PC71BM/ZnO/n-PEDOT:PSS/ PTB7-Th:PC71BM/ETL-1/Al gives a high PCE of 11.47% with $V_{\rm oc}$ = 1.689 V, $J_{\rm sc}$ = 10.51 mA cm^-2, and FF = 64.6%, which is among the highest reported values for tandem OSCs^[12–14,25,37,46] and demonstrates a significant enhancement compared to

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Figure 2. a) The J–V characteristics of tandem OSC and reference subcells under illumination of AM 1.5G (100 mW cm⁻²) and b) EQE spectra of the corresponding subcells.

Table 1. The photovoltaic performance parameters of reference subcells and tandem OSCs. The best PCEs are provided in parentheses.

Device	Active layer thickness [nm]	V _{oc} [V]	$J_{\rm sc}$ [mA cm ⁻²]	FF [%]	PCE [%]
Front cell	90	$\textbf{0.910} \pm \textbf{0.005}$	13.07 ± 0.12	74.0 ± 0.1	8.76 (8.93)
Rear cell	85	0.795 ± 0.005	15.84 ± 0.24	67.8 ± 0.2	8.53 (8.75)
Tandem cell	-	1.684 ± 0.005	10.30 ± 0.21	64.3 ± 0.3	11.21 (11.47)

each corresponding single-junction cells. It should be noted that so far the best reported two-junction tandem cells had a PCE of 11.3%, with a $V_{oc} = 1.54$ V, $J_{sc} = 11.11$ mA cm⁻², and FF = 66%.^[12] So the high PCE shown here was attributed to its higher V_{oc} , which is associated with the high V_{oc} provided by the front cell using the SM as the active materials. It should be noted that the J_{sc} can be improved since the absorption of the polymer used in the rear subcells still have a rather large overlap with that of the SMs used in the front subcells.

EQE measurements of the front subcell and the rear subcell in the tandem device were obtained using 700 nm high-pass and 500 nm low-pass optical filters, respectively.^[47] The EQE measurements show that the front and rear subcells have high EQE values with the maxima of 62% and 57%, respectively, as shown in Figure 2b. This can be attributed to the capability of SMs to utilize the high-energy portion of the solar irradiation efficiently and enable balanced photocurrent from both front and rear cells. Indeed, integrating the EQE with the AM 1.5G solar spectrum shows highly balanced current generation in each subcell with current density of 9.94 mA cm⁻² for the front subcell and 9.25 mA cm⁻² for the rear subcell. The EQE-integrated current densities of the subcells are consistent with the measured $J_{\rm sc}$ of the tandem solar cell, corroborating the accuracy and consistency of our measurements.

In the tandem OSCs with a series connection, the layer thicknesses of the front and rear cells have to be balanced to adjust the current extracted from each layer.^[48] To simplify the thickness optimization process, the active layer thickness of PTB7-Th-based rear cell was optimized to be 85 nm for single-junction cells. A series of tandem cells with different layer thickness for front cells were fabricated under the optimized processing conditions discussed above. The *J*–*V* curves of the tandem cells with different layer thickness of front cells are shown in **Figure 3** and the photovoltaic parameters are summarized in **Table 2**. From Table 2, we can see that V_{oc} remains



Figure 3. The J-V characteristics of the tandem OSCs with different front layer thickness.

almost constant with increasing the layer thickness, and the $J_{\rm sc}$ initially increased and then decreased, indicating that when the front cell thickness was over 90 nm the absorption of the front cell might be too strong to leave sufficient light for generating a symmetric current density in the rear cell. This is consistent with the EQE result (Figure S4, Supporting Information).

 $\label{eq:table 2. The photovoltaic performance parameters of tandem OSCs with different front-layer thicknes.$

Thickness [nm]	V _{oc} [V]	J _{sc} [mA cm ⁻²]	FF [%]	PCE [%]
65	1.681 ± 0.005	10.10 ± 0.15	58.3 ± 0.3	9.55 (10.05)
90	1.684 ± 0.005	10.30 ± 0.21	64.3 ± 0.3	11.21 (11.47)
120	1.669 ± 0.003	9.94 ± 0.15	63.7 ± 0.3	10.57 (10.96)

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Figure 4. The J-V characteristics of the tandem OSCs using DR3TBDTT and DRBDT-TT as the front cell donors and their corresponding subcells.

Using the same strategy and structure, the other two SMs (DR3TBDTT and DRBDT-TT) were also applied as the front cell donor materials, which have an absorption similar to DR3TSBDT.^[40,41] The *J*–*V* curves of the tandem solar cells using the other two SMs are shown in **Figure 4** and the photovoltaic parameters are presented in **Table 3**. The photovoltaic performance parameters of DR3TBDTT and DRBDT-TT based single-junction devices where CuSCN was used as the hole transport layer are shown in Table S3 (Supporting Information). The tandem cells based on these two SMs give high PCEs of 10.73% and 10.43%, respectively, under the same conditions, which also demonstrates the great enhancement compared with their single junction cells. Again, the $V_{\rm oc}$ of ≈1.65 V and $J_{\rm sc}$ of ≈10.5 mA cm⁻² are obtained among the highest literature values, and the overall performance is also among the best compared with literatures.^[12–14,25,37]

In conclusion, three SMs (DR3TSBDT, DR3TBDTT, and DRBDT-TT) were used as the front cell donor materials for highly efficient tandem OSCs which ensured both high Voc and Jsc. The tandem OSCs based on DR3TSBDT prepared under optimal conditions shows a PCE of 11.47%, with a high $V_{\rm oc}$ = 1.689 V, $J_{\rm sc}$ = 10.51 mA cm⁻² and FF = 64.6%, which is among the highest values reported for tandem cells. The tandem OSCs based on DR3TBDTT and DRBDT-TT also showed high PCEs of 10.73% and 10.43%, respectively. However, the overall V_{oc} is a little lower than the sum of V_{oc} of the subcells, suggesting a suboptimal contact at active layer/intermediate layer interface. A higher PCE would be obtained if the ICLs would be further optimized. The same improvement should also be expected if the absoprtion of the active materials in both subcells is optimized and better complementary. All these demonstrate that the monodisperse SMs could perform as promising donor materials for high-performance tandem

Table 3. The photovoltaic performance parameters of reference subcellsand tandem OSC. The best PCEs are provided in parentheses.

Device	V _{oc} [V]	J _{sc} [mA cm ⁻²]	FF [%]	PCE [%]
Ref. subcell ^{a)}	$\textbf{0.881} \pm \textbf{0.005}$	13.84 ± 0.12	73.5 ± 0.3	9.00(9.12)
Tandem cell ^{a)}	1.643 ± 0.005	10.30 ± 0.20	61.5 ± 0.3	10.52(10.73)
Ref. subcell ^{b)}	$\textbf{0.910} \pm \textbf{0.010}$	12.93 ± 0.17	71.0 ± 0.1	8.50(8.70)
Tandem cell ^{b)}	1.671 ± 0.005	10.24 ± 0.14	59.6 ± 0.2	10.28(10.43)

^{a)}DR3TBDTT-based single-junction subcell or tandem solar cell;^{[40] b)}DRBDT-TTbased single-junction subcell or tandem solar cell.^[41] solar cells. Considering the paucity of studies on these small molecules for tandem cells, it should be possible to achieve further improvements and higher performance soon.

Experimental Section

Materials: PTB7-Th (also named as PCE10) was purchased from 1-Material Chemscitech Inc. (St-Laurent, Quebec, Canada) and used as received. CuSCN and PH neutral PEDOT:PSS (n-PEDOT:PSS) were purchased from Sigma–Aldrich Corporation. SMs were synthesized according to previous report.^[39–41] ETL-1 was purchased from Lumtec. ZnO nanoparticles were synthesized following the literature.^[44]

Single-Junction Cell Device Fabrication: The SM:PC71BM single-junction cell was fabricated with a structure of ITO/CuSCN/SM:PC71BM/ETL-1/ Al. A thin layer (30 nm) of CuSCN processed from dimethyl sulfide solution was spin-cast on top of precleaned ITO substrates and annealed in air at 120 °C for 10 min. The active layer was processed according to previous report.^[39-41] For DR3TBDTT:PC71BM, chloroform was used for solvent vapor annealing. The PTB7-Th-based single-device structure was ITO/PEDOT:PSS/PTB7-Th:PC71BM/ETL-1/Al. A thin layer (40 nm) of PEDOT:PSS (Clevios PVP Al 4083) was spin-cast on top of precleaned ITO substrates and annealed in air at 150 °C for 10 min. PTB7-Th:PC71BM was processed with the optimized condition of spin-coating a mixed solvent of choroform/1,8-diiodoctane (98.5%:1.5% by volume) solution (with a total donor and acceptor concentration of 12.5 mg mL⁻¹ and donor/acceptor weight ratio of 1:1.5) at 500 rpm 1.5 s and 1700 rpm for 20 s. Then ETL-1 dissolved in ethanol was spin-coated. The devices were completed after deposition of 60 nm Al as cathode ($\approx 4 \text{ mm}^2$)

Tandem Cell Device Fabrication: The device structure of the tandem cell is shown in Figure 1d. The SM:PC₇₁BM active layers were fabricated via the same process as single-junction cells. The ZnO nanoparticles solution was spin-coated on top of the active layer of the bottom cell, and then n-PEDOT:PSS (around 25 nm) were spin-coated. The PTB7-Th:PC₇₁BM active layers were fabricated via the same process as single-junction cells with thicknesses of 85 nm. The devices were completed after deposition of 60 nm Al as cathode ($\approx 4 \text{ mm}^2$).

Device Characterization: The current density-voltage (J-V) curves of photovoltaic devices were obtained by a Keithley 2400 source-measure unit. The photocurrent was measured under illumination simulated 100 mW cm⁻² AM 1.5G irradiation using a SAN-EI XES-70S1solar simulator, calibrated with a standard Si solar cell. The average PCE was obtained using 50 devices under the same conditions. External quantum efficiencies were measured using Beijing Sevenstar EQE measurement system. The thickness of the active layers in the photovoltaic devices was measured on a Veeco Dektak 150 profilometer.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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