Organic Solar Cells



High-Performance All-Small-Molecule Solar Cells Based on a New Type of Small Molecule Acceptors with Chlorinated End Groups

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While a wide variety of nonfullerene acceptors are developed and perform well in combination with polymer donors, only a few nonfullerene acceptors can work well with small molecule donors. Here, all-small-molecule solar cells with high performance enabled by a new type of small molecule acceptors (F-0Cl, F-1Cl, and F-2Cl), which contain linear alkyl side chains and end groups substituted with various number of chlorine atoms, are reported. End group chlorination leads to redshifted absorption, enhanced crystallinity, and high electron mobility. These properties make them competitive as electron acceptors for all-small-molecule solar devices. When combined with two popular small molecule donors DRTB-T and DRCN5T, these nonfullerene acceptors offer power conversion efficiencies up to 10.76 and 9.89%, which are among the top efficiencies reported in all-small-molecule solar cells and indicate the great potential of all-small-molecule solar devices.

Organic solar cells (OSCs) with bulk heterojunction (BHJ) architecture are considered as a promising solar technology due to the advantages, such as lightweight, low cost, flexibility, etc.^[1-3] The typical BHI active layer of the OSCs consists of a pair of matched materials which function as electron donor and electron acceptor, respectively.^[4] For a long time, the electron acceptor materials have been dominated by fullerene derivatives and power conversion efficiencies (PCEs) of over 11% have been achieved.^[5-11] However, fullerene derivatives suffer from high preparation cost, weak absorptions, and hard tunable energy levels.^[12] Recent years, nonfullerene acceptors (NFAs), especially those acceptor-donor-acceptor (A-D-A) type small molecule NFAs, with advantages of good light absorption, facile synthesis, and fine-tuned energy levels, have drawn great attentions and PCEs over 14%^[13-17] have achieved through the materials innovation, morphology control, and device optimization.[18-23]

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On the other hand, the electron donor materials could generally be divided into two categories: polymers and small molecules. Both types of donors have showed high performance with PCE over 11% in combination with fullerene acceptors.^[6,8] Nevertheless, for NFA-based OSCs with high efficiencies, most donor materials are wide-bandgap polymers^[24-27] and only a few small molecule donors^[28-31] work well with several very limited NFAs. In consideration of the unique advantages of small molecule materials.^[32,33] such as well-defined structure and thus less batch-to-batch variation, versatile chemical structures and thus easier energy level control, much efforts have been made to develop all-small-molecule (ASM) OSCs,

which comprise small molecule donor and nonfullerene small molecule acceptor. Presently, promising PCEs with value over 10% have been achieved by Li and Hou's groups.^[30,34,35] But so far, the electron acceptors with decent performance used in the reported ASM-OSCs have only been found in the case of (2,2'-[(4,4,9,9-tetrahexyl-4,9-dihydro-sindaceno[1,2-*b*:5,6-*b*'] dithiophene-2,7- diyl)bis[methylidyne(3-oxo-1*H*-indene-2,1(3H)-diylidene)]]bis-propanedi-nitrile) (IDIC).^[36] Considering the versatile chemical structures and thus easier energy level control of small molecules for OSCs, it is highly possible and even better high performing acceptors could be designed and developed for ASM-OSCs.

Recently, our group has reported an A-D-A type small molecule acceptor, 2,9-bis-(2methylene(3(1,1dicyanomethylene)indanone))7,12-dihydro-4,4,7,7,12,12-hexaoctyl-4Hcyclopenta[2",1":5,6;3",4":5',6']-di-indeno[1,2-b:1',2'-b'] dithiophene (FDICTF, F-0Cl in Figure 1) and PCE over 10% has been achieved using wide-bandgap polymer PBDB-T as the donor.^[37] To further tune its band structure and morphology in active layer, another acceptor F-1Cl through introduction of one chlorine atom on the end groups of F-0Cl was developed and a PCE over 11% was obtained for the devices based on PBDB-T:F-1Cl, which was attributed to the redshifted absorption, enhanced molecular packing, and electron mobility of F-1Cl.^[38] Encouraged by the success of polymer donor devices, acceptor F-1Cl might be a good choice for ASM OSCs. Just recently, it has been reported that acceptors with high crystallinity are favorable in ASM-OSCs.^[39-41] Whether the crystallinity of F-1Cl

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Figure 1. a) The chemical structures of the acceptors (F-0Cl, F-1Cl, and F-2Cl). b) The chemical structure of the donor DRCN5T. c) The UV-vis absorption of these three acceptors in solid films. d) Energy-level diagrams of the donor and acceptor materials.

acceptor is optimal and how the crystallinity of acceptors affects their properties of ASM-OSCs need to be comprehensively investigated. With this content, two Cl atoms were introduced and another acceptor F-2Cl (Figure 1) was synthesized. Thus in this work, we wish to report ASM OSCs using three small molecule F-nCl (n = 0, 1, 2) as acceptors (Figure 1) with two well-known small molecule donors,^[28,42] and study the impact of introduction of Cl atom in the same main molecule structure. After device optimization, PCEs of 5.49, 8.12, and 9.89% were achieved for the F-0Cl, F-1Cl, and F-2Cl based devices, respectively, when using DRCN5T as the small molecule donor. The high crystallinity of F-2Cl gave rise to its high device performances. With this, DRTB-T was selected as the donor and an impressive PCE of 10.76% was achieved for the F-2Cl-based devices. These results demonstrate that F-2Cl is a promising NFA molecule for ASM OSCs and synergistic investigation of donor and acceptor molecules is needed to obtain highperformance ASM OSCs.

The synthesis of F-2Cl is shown in the Supporting Information. The dialdehyde intermediate and two chlorine atoms substituted end units were prepared according to our previous work and the reported literatures.^[37] The UV–vis absorption spectra of the three small molecule acceptors in solid films are shown in Figure 1c. F-*n*Cl (n = 0, 1, 2) films display similar absorption profile. The introduction of chlorine atoms not only enhanced the absorption spectra. Compared to their absorption spectra in dilute chloroform solution (Figure S4, Supporting Information), the absorption peaks are redshifted by 24 nm for F-0Cl, 33 nm for F-1Cl, and 41 nm for F-2Cl, respectively. The absorption edges of F-*n*Cl (n = 0, 1, 2) films are 760, 786,

and 805 nm, and thus their corresponding optical bandgaps are 1.63, 1.58, and 1.54 eV, respectively.

The molecular energy level of the donor DRCN5T and the three acceptors were measured via cyclic voltammetry and the results are illustrated in Figure 1d. The lowest unoccupied molecular orbital (LUMO) levels of F-*n*Cl (n = 0, 1, 2) are estimated to be -3.71, -3.75, and -3.86 eV, respectively. The highest occupied molecular orbital (HOMO) levels are -5.43, -5.46, and -5.50 eV, respectively. With the number of chlorine atoms increasing, the LUMO and HOMO levels all shifted down, especially the LUMO levels. As seen in Figure 1c, each combination of the DRCN5T donor and the F-*n*Cl (n = 0, 1, 2) acceptor provides sufficient energy offsets for efficient exciton dissociation.

To ascertain the effect of chlorination on intermolecular packing, we characterized the neat acceptor films via grazingincidence wide-angle X-ray scattering (GIWAXS). The pristine F-0Cl film (**Figure 2**a) shows a very weak (010) reflection of π – π stacking in the out-of-plane (OOP) direction, indicative of disordered packing. After the introduction of chlorine atoms to the end group, F-1Cl and F-2Cl neat films exhibit well-defined (010) reflection peak in OOP direction together with the appearance of in-plane (IP) (100) peaks, indicating preferential faceon orientation. The (010) reflection peak of F-*n*Cl (n = 0, 1, 2) are located at 1.77, 1.82, and 1.84 Å⁻¹, respectively, and thus the corresponding π - π stacking distances are 3.55, 3.45, and 3.41 Å, respectively, revealing that chlorination led to a more condensed stacking in the π - π stacking direction with higher intensity. This result was consistent with the redshifted absorption spectrum of their thin films. In addition, we found that the π - π coherence lengths, calculated from the full-width at half-maximum of the OOP (010) reflection peak, were 0.59, 5.67,



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Figure 2. GIWAXS patterns of the pure film of F-0Cl, F-1Cl, and F-2Cl.

and 6.43 nm for F-0Cl, F-1Cl, and F-2Cl, respectively, which demonstrated that the crystallinity of molecules increased with introduction of chlorine atoms on the end groups.

All-small-molecule OSCs with the conventional configuration of indium tin oxide (ITO)/poly(3,4-ethylenedioxythiopene): poly(styrenesulfonate) (PEDOT:PSS)/DRCN5T:acceptor/perylene diimide functionalized with amino N-oxide (PDINO)/Al were adopted to evaluate the photovoltaic performance of these three small molecule acceptors. Solvent vapor and thermal annealing treatment were used to optimize the performance, and the optimization details are described in Tables S1–S10 (Supporting Information). It can be found that the best performance of DRCN5T:F-0Cl/F-1Cl/F-2Cl based devices could be obtained via solvent vapor annealing with CH_2Cl_2 for 40 s, thermal annealing at 110 °C for 10 min, and thermal annealing at 110 °C for 8 min, respectively. As a result, the active layers based on DRCN5T:F-0Cl/F-1Cl/ F-2Cl can obtain the PCEs of 5.49, 8.12, and 9.89%, respectively.

The optimized current density versus voltage (J-V) and external quantum efficiency (EQE) curves of the OSCs based on these three blends are presented in **Figure 3**a,b, respectively, and the corresponding parameters are listed in **Table 1**. As can



Figure 3. a) Current density–voltage (*J*–*V*) curves, b) EQE curves, c) photocurrent density versus effective voltage ($J_{ph} - V_{eff}$), and d) double logarithmic plots of J_{sc} as a function of incident light intensity for the optimal devices based on DRCN5T:F-0Cl, DRCN5T:F-1Cl, and DRCN5T:F-2Cl.

 Table 1. Photovoltaic parameters for the DRCN5T:F-0Cl, DRCN5T:F-1Cl, and DRCN5T:F-2Cl-based solar cells.

Acceptors	$V_{\rm oc} [V]$	J _{sc} [mA cm ⁻²]	FF [%]	PCE [avg.] ^{a)} [%]	$J_{\rm sc}^{\rm b)}$ [mA cm ⁻²]
F-0Cl	1.090	9.64	52.3	5.49 (5.17)	9.49
F-1Cl	0.975	13.07	63.7	8.12 (7.90)	12.99
F-2Cl	0.906	15.97	68.4	9.89 (9.62)	15.68

 $^{\rm a)} \rm Average$ PCE values obtained from ten devices are shown in parentheses; $^{\rm b)} \rm The}$ $J_{\rm sc}$ calculated from the EQE spectrum.

be seen, the V_{oc} decreased sharply from 1.090 V of F-0Cl to 0.975 V of F-1Cl and to 0.906 V of F-2Cl, which is consistent with the downshifted LUMO levels of the acceptors. On the contrary, the J_{sc} and FF increased after introduction of chlorine atoms. Thus, the PCE of the chlorinated acceptors based device improved significantly. F-2Cl-based all-small-molecule solar cells gave the highest J_{sc} of 15.97 mA cm⁻² and FF of 0.684. In Figure 3b, the EQE spectra are broadened and enhanced gradually from F-0Cl to F-1Cl and F-2Cl, resembling the absorption of the three acceptors (Figure 1c). The maximum EQE values of F-0Cl, F-1Cl, and F-2Cl based devices are 45.5, 59.3, and 67.9%, respectively. The J_{sc} values of F-0Cl, F-1Cl, and F-2Cl-based devices are 9.49, 12.99, and 15.68 mA cm⁻², respectively, which are in good agreement with the J_{sc} values obtained from the J-V curves.

To further probe the reason of the enhanced $J_{\rm sc}$ and FF by chlorination, the relation of photocurrent density ($J_{\rm ph} = J_{\rm L} - J_{\rm D}$, where $J_{\rm L}$ and $J_{\rm D}$ are the light and dark current density, respectively) versus effective voltage ($V_{\rm eff} = V_0 - V_{\rm a}$, where V_0 is the voltage at $J_{\rm ph} = 0$ and $V_{\rm a}$ is the applied voltage) are plotted in Figure 3c.^[43] Under short-circuit conditions, the $J_{\rm ph}/J_{\rm ph,sat}$

values for F-1Cl and F-2Cl-based devices were 94.7 and 97.9%, respectively, manifesting the effective exciton dissociation and charge collection efficiency. Under maximal power output circumstances, the ratios of $J_{ph}/J_{ph,sat}$ for the chlorinated acceptors (F-1Cl and F-2Cl) were 77.4 and 83.6%, respectively, suggesting a considerably higher charge collection efficiency via end group chlorination. The lowest $J_{ph}/J_{ph,sat}$ value of 76.6% under short-circuit condition and 53.3% under maximal power output circumstances were obtained in the F-0Cl-based device, which is in accordance with its lowest EQE values. These results demonstrate that the end group chlorination of acceptors in all-small-molecule OSCs is an effective approach to improve the exciton dissociation and overall charge collection, and thus improving J_{sc} and FF simultaneously.^[44,45] The J_{sc} under different light intensities (P) was also measured. As presented in Figure 3d, I_{sc} and P follow the relationship of $I_{sc} \propto P^{\alpha}$.^[46] With the number of chlorine atom increasing, α of the fitting curves increased (0.88 for F-0Cl, 0.94 for F-1Cl, and 0.99 for F-2Cl). This means that the bimolecular recombination could be reduced for the chlorinated molecules based devices and their FFs thus were enhanced.

Since the morphology of active layer is critical for device performance, we utilized atomic force microscopy (AFM) and transmission electron microscopy (TEM) to detect the three optimized blend films based on DRCN5T and F-*n*Cl. Figure 4 shows the AFM and TEM images of the three blend films. The active layers of DRCN5T:F-0Cl, DRCN5T:F-1Cl, and DRCN5T:F-2Cl display very uniform and smooth surface with small rootmean-square (RMS) roughness of 1.44, 1.52, and 1.45 nm, respectively. The domain size increased after chlorination. From TEM images (Figure 4), indistinct nanostructures could be found in F-0Cl-based blend film; after chlorination of end groups, clear



Figure 4. Tapping-mode AFM height images and TEM images of the optimal active layers based on a,d) DRCN5T:F-0Cl, b,e) DRCN5T:F-1Cl, and c,f) DRCN5T:F-2Cl. The scale bars are 200 nm.

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Figure 5. a) The chemical structure of DRTB-T, b) the J-V curve, and c) EQE for the optimal devices based on DRTB-T:F-2Cl.

bicontinuous interpenetrating network was observed, beneficial for the charge transport and thus improved J_{sc} and FF.

The morphology variation of the active layers was further manifested by the results of charge transport properties of the blend films (Figure S8, Supporting Information). The carrier mobilities of the blend films were measured by space-charge limited current (SCLC) measurement.^[47] The electron mobilities (μ_e) of F-0Cl, F-1Cl, and F-2Cl based blend films were 0.56 × 10⁻⁴, 1.62 × 10⁻⁴, and 2.22 × 10⁻⁴ cm² V⁻¹ s⁻², respectively. The increase of electron mobility with chlorination was closely related to the enhanced face-on packing of the acceptors as discussed above. The similar trend on hole mobility (μ_h) of these three blends was obtained with values of 1.45 × 10⁻⁴, 2.04 × 10⁻⁴, and 2.50 × 10⁻⁴ cm² V⁻¹ s⁻², respectively. F-2Cl-based device showed the highest hole and electron mobilities and the most balanced charge transport ($\mu_h/\mu_e = 1.12$), which is very important to obtain higher PCE.

From the above results, small molecule F-2Cl is a promising acceptor for fabricating all-small-molecule solar cells due to its redshifted absorption spectrum, enhanced crystalline and high electron mobility. Recently, small molecule DRTB-T (Figure 5a) has been used as donor to fabricate ASM OSCs and gave promising performances with IDIC.^[28] Considering the above results, we fabricated ASM OSC with DRTB-T as donor and F-2Cl as acceptor. After device optimization, a PCE of 10.76% was achieved (Figure 5b,c). The spectra of the small molecule donor DRTB-R possess a strong and broad absorption in the region of 300-600 nm,^[28] more complementary to that of F-2Cl film, which is beneficial to utilize the visible region (300-800 nm) of sunlight, and thus higher J_{sc} (17.24 mA cm⁻²) than that of DRCN5T:F-2Cl based device. DRTB-T:F-2Cl-based device achieved much higher V_{oc} (0.969 V), originating from the lower HOMO level of the donor material. However, the relatively low exciton dissociation and charge collection efficiency as displayed in J_{ph}-V_{eff} characteristic plot (Figure S8, Supporting Information, the $J_{\rm ph}/J_{\rm ph,sat}$ value is 93.2% under short-circuit condition and 72.9% under maximal power output circumstances) should be responsible for the slightly low FF of DRTB-T-based device.^[48]

In summary, ASM OSCs employing three nonfullerene acceptors (F-0Cl, F-1Cl, and F-2Cl) were investigated. It has been found that F-2Cl with high crystallinity gave best PCE of 9.89% when DRCN5T was used as donor molecule. An impressive PCE of 10.76% has been achieved for the DRTB-T:F-2Cl-based

devices. Our results indicated that increasing the crystallinity of the acceptor through chlorination and other molecule engineer is an effective strategy to obtain high-efficiency ASM OSCs. Meanwhile, donor molecules should also be taken into consideration synergistically from perspective of absorption, energy levels, etc., matched with acceptors.

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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