

Cyano-Functionalized Quinoxaline-Based Polymer Acceptors for All-Polymer Solar Cells and Organic Transistors

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Quinoxaline (Qx) derivatives are promising building units for efficient photovoltaic polymers owing to their strong light absorption and high charge-transport abilities, but they have been used exclusively in the construction of polymer donors. Herein, for the first time, Qx-based polymer acceptors (P_A s) were developed by introducing electron-withdrawing cyano (CN) groups into the Qx moiety (QxCN). A series of QxCN-based P_A s, P(QxCN-T2), P(QxCN-TVT), and P(QxCN-T3), were synthesized by copolymerizing the QxCN unit with bithiophene, (*E*)-1,2-di-(thiophene-2-yl)ethene, and terthiophene, respectively. All of

Introduction

All-polymer solar cells (all-PSCs) comprising polymer donor (P_D) and polymer acceptor (P_A) have been intensively studied owing to their superior mechanical and thermal stabilities.^[1] Recently, a significant improvement in their power conversion efficiency (PCE) to over 14% has been achieved by the development of novel P_{AS} that polymerize small molecular acceptors (SMA- P_{AS}) with high light absorption coefficients and good electron transport abilities.^[2] However, the molecular weights of SMA-P_As are typically far lower than the critical molecular weight. Therefore, the intrinsic mechanical and thermal properties of SMA-P_A-based all-PSCs still remain poor, which are comparable to those of SMA-based PSCs.^[3] Among the several P_A candidates, naphthalenediimide (NDI)-based P_As (e.g., N2200) are still dominantly utilized owing to their desirable semi-crystalline properties and the synthetic availability to achieve the high molecular weight, which endows the all-PSCs with superior mechanical and thermal reliabilities.^[1d,4]

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the $P_{\rm A}$ s exhibited unipolar n-type characteristics with organic field-effect transistor (OFET) mobilities of around 10^{-2} cm²V⁻¹s⁻¹. In space-charge-limited current devices, P-(QxCN-T2) and P(QxCN-TVT) exhibited electron mobilities greater than 1.0×10^{-4} cm²V⁻¹s⁻¹, due to the well-ordered structure with tight π - π stacking. When the $P_{\rm A}$ s were applied in all-polymer solar cells (all-PSCs), the highest performance of 5.32% was achieved in the P(QxCN-T2)-based device. These results demonstrate the significant potential of Qx-based $P_{\rm A}$ s for high-performance all-PSCs and OFETs.

Most NDI-based P_As, however, have low absorption coefficients, which limit the overall exciton generation in all-PSCs.^[1e,3c,5] Moreover, the NDI building block has a small room for structural tunability, which makes further enhancement of the optical and electrical properties of NDI-based P_As challenging. Accordingly, extensive efforts to develop new n-type building blocks such as double boron-nitrogen coordinated bipyridine (BNBP) and cyano-substituted benzothiadiazole (DCNBT) have been made, and the resulting P_{AS} have produced all-PSCs with PCEs exceeding 10%.[1c,5a,6] Nevertheless, in comparison to a large number of p-type building blocks for efficient $P_{D}s$, there is still a lack of promising electron-deficient building blocks available for constructing new $P_{A}s$.^[1c] Thus, the design of new n-type building blocks is imperative to broaden the efficient P_A library and further improve the device performance and stability of all-PSCs.

In this regard, we pay our attention to quinoxaline (Qx)based unit as a building block for developing efficient $P_{A}s$ due to the following reasons: (i) Qx structure intrinsically has electron-deficient nature originated from electron-withdrawing nitrogen atoms embedded in its conjugated system.^[7] (ii) The desirable backbone planarity of the Qx structure induced by intra- and inter-molecular interactions can provide both high absorption coefficients and efficient charge-transport abilities in Qx-based polymers.^[8] (iii) The Qx unit can be easily functionalized. For example, various electronegative substituents can be introduced in the 6- and 7-positions.^[9] It is noted that this synthetic accessibility of the Qx unit facilitates tuning of its electron deficiency and frontier molecular orbital energy levels. Therefore, we envision that these key features make the Qx derivatives attractive for developing efficient P_{As} . However, to the best of our knowledge, Qx-based P_{AS} have not yet been reported so far.



Herein, we report the development of a series of new P_{AS} based on cyano (CN)-functionalized Qx derivative (QxCN). We incorporate CN groups into the 6- and 7-positions of the Qx moiety to produce QxCN that is sufficiently electron-deficient for use in building $P_{A}s$. In particular, the use of the CN group lowers the lowest unoccupied molecular orbital energy level (E_{LUMO}) of the Qx unit to a level that is suitable for efficient electron injection and transport.^[10] In addition, the CN group has a large dipole moment, which facilitates charge separation at the donor-acceptor interfaces of the active layer by reducing Coulomb attraction and exciton binding energy.^[5a,11] To tune the structural and electrical properties of QxCN-based P_A s, three different electron-donating units of bithiophene (T2), (E)-1,2di(thiophen-2-yl)ethene (TVT), and terthiophene (T3) are copolymerized with the QxCN unit to produce poly[(2,3-bis(3,4-bis (octyloxy)phenyl)quinoxaline-6,7-dicarbonitrile-5,8-diyl)-alt-5,5'-(2,2'-bithiophene)] (P(QxCN-T2)), poly[(2,3-bis(3,4-bis(octyloxy) phenyl)quinoxaline-6,7-dicarbonitrile-5,8-diyl)-alt-5,5'-((E)-1,2-bis (thiophen-2-yl)ethene)] (P(QxCN-TVT)), and poly[(2,3-bis(3,4-bis (octyloxy)phenyl)quinoxaline-6,7-dicarbonitrile-5,8-diyl)-alt-5,5"-(2,2':5',2"-terthiophene)] (P(QxCN-T3)), respectively. All the QxCN-based P_As exhibit sufficiently high absorption coefficients of $(4.3-5.3) \times 10^4$ cm⁻¹. In addition, all of the QxCN-based P_As have unipolar n-type characteristics with organic field-effect transistor (OFET) mobility ($\mu_{\mathrm{e,OFET}}$) values of around 10⁻² cm²V⁻¹s⁻¹. In space-charge-limited current (SCLC) devices, the reasonably high SCLC electron mobility ($\mu_{e,SCLC}$) values of 2.0×10^{-4} and $3.8 \times 10^{-4} \, \text{cm}^2 V^{-1} \text{s}^{-1}$ are observed in the P(QxCN-T2) and P(QxCN-TVT) polymers, respectively. Then, the three P_{AS} are blended with poly[(2,6-(4,8-bis(5-(2-ethylhexyl)thiophen-2yl)-benzo[1,2-b:4,5-b']dithiophene))-alt-(5,5-(1',3'-di-2-thienyl-5',7'-bis(2-ethylhexyl)benzo[1',2'-c:4',5'-c']dithiophene-4,8-dione))] (PBDB-T) to produce all-PSCs. The highest performance of all-PSCs (PCE = 5.32%) is achieved by the P(QxCN-T2):PBDB-T blend. Our results demonstrate that the QxCN unit can be a new promising building block for P_As applicable in all-PSCs and OFETs.

Results and Discussion

To develop a series of Qx-based P_A s, we first functionalized the 6- and 7-positions of the Qx unit with CN groups to produce the QxCN unit. The strong electron-withdrawing ability of CN groups effectively decreases E_{LUMO} of conjugated materials to enable efficient electron injection and transport.^[10] The QxCN monomer (3) was synthesized through a coupling reaction between 1,2-bis(3,4-bis(octyloxy)phenyl)ethane-1,2-dione (1) and 4,5-diamino-3,6-dibromophthalonitrile (2) using a modified synthesis procedure previously reported (Figure 1a).^[9a] To systematically control the electrical and structural properties of QxCN-based P_As, three different electron-rich units (T2, TVT, and T3) were selected and copolymerized via Stille coupling with the QxCN unit to produce P(QxCN-T2), P(QxCN-TVT), and P(QxCN-T3), respectively (Figure 1b). The detailed synthesis procedures and results are described in the Experimental Section and Supporting Information (Figure S1). Figure 2 shows thin-film UV/Vis absorption spectra and energy level alignment diagram of the resulting P(QxCN-T2), P(QxCN-TVT), and P(QxCN-T3) P_{A} s and PBDB-T P_{D} . PBDB-T was selected as the P_{D} in the all-PSCs because of the good complementary light absorption property and proper energy level alignment with the QxCNbased P_{AS} .^[12] The basic characteristics of the three P_{AS} and the P_D including molecular weight, electrochemical, and optical properties are summarized in Table 1. All the synthesized P_{AS} had reasonably high number-average molecular weight (M_n) (over 90 kg mol⁻¹) and similar dispersity (D) values (Figure S2). In UV/Vis spectra of thin-films, while the absorption profile of P(QxCN-T3) was slightly red-shifted relative to those of the other P_As, all the QxCN-based P_As mainly absorbed light at wavelength near 400 and 550-800 nm with sufficiently high absorption coefficients in the range of $(4.3-5.3) \times 10^4$ cm⁻¹ (Figure 2a). To investigate the electrochemical properties of the QxCN-based P_{AS} , the highest occupied molecular orbital energy level ($E_{\rm HOMO}$) and $E_{\rm LUMO}$ of each $P_{\rm A}$ were determined from the onset of oxidation and reduction potentials, respectively, by measuring the cyclic voltammetry (CV) (Figure S3). The $E_{\rm HOMO}/$ *E*_{LUMO} of P(QxCN-T2), P(QxCN-TVT), and P(QxCN-T3) were determined to be -5.73/-3.58 eV, -5.71/-3.56 eV, and -5.68/



Figure 1. (a) Synthesis route for QxCN monomer and (b) chemical structures of three QxCN-based P_As and a PBDB-T P_D.

ChemSusChem 2021, 14, 1-9 www.chemsuschem.org 2 These are not the final page numbers!





Figure 2. (a) UV/Vis absorption spectra of thin films and (b) frontier molecular orbital energy levels of three QxCN-based P_As, P(QxCN-T2), P(QxCN-TVT), and P(QxCN-T3), and P_D, PBDB-T.

-3.62 eV, respectively. Compared to the E_{LUMO} of typical NDIbased P_{A} s (e.g., E_{LUMO} of N2200 = -4.02 eV),^[13] the high-lying E_{LUMO} s of QxCN-based P_{A} s were expected to produce higher open-circuit voltage (V_{OC}) values in all-PSCs than those in the devices with NDI-based P_{A} s. Notably, the slightly narrower bandgap of P(QxCN-T3) agreed well with the decreased optical bandgap ($E_{\text{g}}^{\text{opt}}$), which might be attributed to the more extended conjugation of T3 as compared with T2 and TVT moieties.^[13,14] The three QxCN-based P_{A} s showed 5% weight loss points between 336 and 372 °C in the thermal gravimetric analysis (TGA). No distinct thermal transitions were observed during differential scanning calorimetry (DSC) measurements (Figure S4).

We evaluated the charge-transport characteristics of the QxCN-based P_{AS} by measuring the $\mu_{e,OFET}$ values of OFET devices having top-gate and bottom-contact architecture (Table 2). The detailed procedure for the OFET fabrication is described in the Supporting Information. Interestingly, all of the QxCN- P_{A} -based OFET devices exhibited the unipolar n-type transfer and output characteristics with $\mu_{e,OFET}$ values ranging from 7.4 × 10⁻³ to 2.4 × 10⁻² cm²V⁻¹s⁻¹ (Figure S5). Among them, P(QxCN-TVT) demonstrated the highest $\mu_{e,OFET}$ of 2.4 × 10⁻² cm²V⁻¹s⁻¹, which is

consistent with the trends of previously reported NDI-based P_{AS} containing TVT unit.^[13,15] Thus, the incorporation of CN groups into the Qx moiety successfully enhanced its electron deficiency to afford n-type characteristics in QxCN-based polymers. Then, we fabricated the electron-only SCLC devices to investigate the charge-transport properties in a through-plane direction (Table 2). This vertical transport is an important factor for determining the photovoltaic parameters in all-PSCs. P(QxCN-T2) and P(QxCN-TVT)-based devices showed higher μ_{eSCLC} values of 2.0×10^{-4} and 3.8×10^{-4} cm²V⁻¹s⁻¹, respectively, than P-(QxCN-T3)-based devices $(6.9 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1})$. The superior SCLC mobilities of P(QxCN-T2) and P(QxCN-TVT) were mainly attributed to face-on orientation of the polymer crystals with well-ordered structures and a shorter π - π stacking distance (3.65 Å) than those of P(QxCN-T3) as revealed by grazing incidence wide-angle X-ray scattering (GIWAXS) measurements (Figure 3 and Table S1).^[16] In comparison, the P(QxCN-T3) film exhibited relatively lower crystalline characteristics with a longer π - π stacking distance of 3.76 Å. It might originate from the restricted rotational freedom of T3 group, which often hinders efficient π - π stacking between the conjugated polymers. $^{[8b,17]}$ It is worthwhile to note that the $\mu_{ extsf{e}, extsf{SCLC}}$ values of

Table 1. Material properties of the P(QxCN-T2), P(QxCN-TVT), and P(QxCN-T3) P _A s, and PBDB-T P _D used in this study.									
Polymer	M _n ^[a] [kg mol⁻¹]	${\cal D}^{[a]}$	$\lambda_{\max}^{[b]}$ [nm]	Absorption coefficient ^(b) [cm^{-1}]	E ^{gopt.[b]} [eV]	Е _{номо} ^[с] [eV]	E _{LUMO} ^[c] [eV]		
P(QxCN-T2) P(QxCN-TVT) P(QxCN-T3) PBDB-T	112 176 99 124	4.3 5.3 5.2 2.5	700 687 709 623	4.3×10^4 5.3×10^4 5.1×10^4 6.4×10^4	1.51 1.52 1.48 1.83	5.73 5.71 5.68 5.57	-3.58 -3.56 -3.62 -3.42		

[a] Determined by gel permeation chromatography (GPC) with 80 °C heated 1,2-dichlorobenzene. [b] Determined from the UV/Vis absorption spectra of thin films. [c] Measured from the CV curves.

Table 2. Electron mobilities measured using OFET and SCLC devices based on QxCN-based P _A s.						
Polymer	OFET $\mu_{e,OFET,avg}^{[a]}$ [cm ² V ⁻¹ s ⁻¹]	I _{on} /I _{off}	V _{th} ^[a] [V]	$\overset{SCLC}{\underset{\mu_{e,SCLC,avg}}{^{[a]}}}[cm^2V^{-1}s^{-1}]$		
P(QxCN-T2) P(QxCN-TVT) P(QxCN-T3)	$(7.6 \pm 0.4) \times 10^{-3}$ $(2.4 \pm 0.1) \times 10^{-2}$ $(7.4 \pm 0.5) \times 10^{-3}$	$> 10^{3}$ > 10^{3} > 10^{3}	$\begin{array}{c} 19.9 \pm 0.7 \\ 23.1 \pm 0.5 \\ 26.7 \pm 0.4 \end{array}$	$(2.0 \pm 0.5) \times 10^{-4}$ $(3.8 \pm 0.2) \times 10^{-4}$ $(6.9 \pm 0.1) \times 10^{-5}$		
[a] The average values were determined from the measurements of more than 10 devices.						

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Figure 3. (a) In-plane and (b) out-of-plane direction line-cut profiles of GIWAXS for pristine films of QxCN-based P_As.



Figure 4. (a) J-V curves and (b) EQE spectra of all-PSCs blended with PBDB-T as P_{D} .

P(QxCN-T2) and P(QxCN-TVT) films, of over 1.0×10^{-4} cm²V⁻¹s⁻¹, are higher than those of representative NDI- and perylenediimide (PDI)-based P_A pristine films.^[1f,4b,11b,12b,18]

To examine the potential of the QxCN-based polymers as electron acceptors, all-PSCs with PBDB-T $P_{\rm D}$ were fabricated with a conventional device structure of indium tin oxide (ITO)/ poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PE-DOT:PSS)/active layer/poly[(9,9-bis(3'-((N,N-dimethyl)-N-ethylammonium)propyl)-2,7-fluorene)-alt-5,5'-bis(2,2'-thiophene)-2,6naphthalene-1,4,5,8-tetracaboxylic-N,N'-di(2-ethylhexyl)imide] dibromide (PNDIT-F3N-Br)/Aq.^[19] The detailed device fabrication condition is described in the Supporting Information. Figure 4a presents the current density - voltage (J-V) characteristics measured under AM 1.5G illumination at 100 mW cm⁻², and the photovoltaic parameters are summarized in Table 3. All the $QxCN-P_A$ -based devices exhibited high V_{OC} values of over 1.0 V that are significantly higher than those of NDI-P_A-based all-PSCs using PBDB-T P_D.^[1h,12,20] The P(QxCN-T2)-based all-PSCs achieved a maximum PCE of 5.32% with a short-circuit current density (J_{SC}) of 10.57 mA cm⁻² and a fill factor (FF) of 0.49. In comparison, the P(QxCN-TVT)-based all-PSCs exhibited a lower PCE of 3.84% ($J_{sc} = 8.48 \text{ mA cm}^{-2}$; FF = 0.45) and the P(QxCN-T3)-based devices had a very low PCE of 0.72%. In addition, the external quantum efficiency (EQE) spectra of the all-PSCs were measured (Figure 4b). The J_{SC} values of the all-PSCs were wellmatched with the corresponding integrated J_{SC} obtained from the EQE spectra within 6% error. The EQE spectrum of the P(QxCN-T2)-based all-PSCs reflected higher responses than those of the other all-PSCs over the entire range of wavelength (350-800 nm). Therefore, the trends of photovoltaic performances were mainly determined by the differences in J_{SC} and FF values.

To understand the photovoltaic trends of all-PSCs, we investigated their charge recombination process by analyzing the dependence of J_{SC} and V_{OC} on light intensity (P) (Figure 5).^[21] It is known that a power-law relationship exists between J_{SC} and *P* ($J_{SC} \propto P^{\alpha}$), and the slope (α) of ln(J_{SC}) versus ln(*P*) is close to unity when bimolecular recombination is negligible during device operation (Figure 5a). Among the all-PSCs, the P(QxCN-T3)-based device had the lowest α value of 0.76, indicating that the severe bimolecular recombination occurs during operation. In contrast, both the P(QxCN-T2)- and P(QxCN-TVT)-based

Table 3. Photovoltaic performances of all-PSCs blended with PBDB-T as $P_{\rm D}$.						
Active layer	<i>V</i> _{OC} ^[a] [V]	$J_{\rm SC}^{[\rm a]} [{\rm mA cm^{-2}}]$	FF ^[a]	$PCE_{avg}^{[a]}$ (PCE_{max}) [%]		
P(QxCN-T2) blend P(QxCN-TVT) blend P(QxCN-T3) blend	$\begin{array}{c} 1.02\pm 0.01 \\ 1.00\pm 0.01 \\ 1.03\pm 0.01 \end{array}$	$\begin{array}{c} 10.25 \pm 0.44 \\ 7.83 \pm 0.95 \\ 2.16 \pm 0.30 \end{array}$	$\begin{array}{c} 0.48 \pm 0.02 \\ 0.45 \pm 0.01 \\ 0.26 \pm 0.02 \end{array}$	$\begin{array}{c} 5.02 \pm 0.26 \; (5.32) \\ 3.54 \pm 0.43 \; (3.84) \\ 0.58 \pm 0.11 \; (0.72) \end{array}$		
[a] The average values were determined from the measurements of more than 10 devices.						

ChemSusChem 2021, 14, 1-9

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Figure 5. Dependence of the (a) J_{sc} and (b) V_{oc} on light intensity (P) in all-PSCs.



Figure 6. (a) Photocurrent analysis of all-PSCs. (b–d) PL spectra of P(QxCN-T2), P(QxCN-TVT), and P(QxCN-T3) pristine films and their blend films with PBDB-T ($\lambda_{excitation} = 750 \text{ nm}$).

devices exhibited relatively higher α values and, in particular, the P(QxCN-T2) blend film showed the highest α value of 0.91. Under the open-circuit condition, the V_{OC} of all-PSCs is proportional to the natural logarithm of $P[V_{OC} \propto \ln(P)]$, and the slope (*S*) with a unit of k_BTq^{-1} (k_B =Boltzmann constant, T=temperature, and q=elementary charge) approaches unity when no monomolecular or trap-assisted recombination occurs. As shown in Figure 5b, the P(QxCN-T2)- and P(QxCN-TVT)-based devices had the *S* values of 1.36–1.38 k_BTq^{-1} , respectively, whereas the *S* value of the P(QxCN-T3)-based device was very high as 2.01 k_BTq^{-1} . These results suggest that trap-assisted or monomolecular recombination is most suppressed in the P(QxCN-T2)-based device. Thus, the very low J_{SC} and FF of the P(QxCN-T3)-based all-PSCs can be explained by the severe charge recombination.^[22]

Next, we investigated the exciton-dissociation probabilities [P(E,T)s] of the all-PSCs by measuring the photocurrent density (J_{ph}) against the effective voltage (V_{eff}) . Figure 6a shows the J_{ph} - V_{eff} curves of the QxCN- P_A -based all-PSCs. The P(E,T) value for each blend was calculated from the ratio between the J_{ph} value under short-circuit condition and saturated photocurrent den-

sity (J_{sat}) at $V_{eff} = 6$ V. The P(QxCN-T2)-based all-PSC had P(E,T) of 72%, while the P(QxCN-TVT)-based all-PSC exhibited P(E,T) of 65%. The higher P(E,T) value of the P(QxCN-T2)-based device indicates better exciton-dissociation behavior, which explains the higher J_{SC} value of the device. In stark contrast, the J_{ph} of the P(QxCN-T3)-based device began to rise over 1 V and it was not saturated even at 6 V with only 29% of P(E,T) value. Thus, the photo-induced excitons in the P(QxCN-T3)-based device were poorly dissociated into free charge carriers. This behavior may be explained by the relatively high number of electrondonating thiophene units in T3 that weakens the electronaccepting ability of P(QxCN-T3), hindering exciton dissociation into holes and electrons at the interfaces between the P(QxCN-T3) P_A and PBDB-T P_D .^[23] To support the trends of P(E,T) in the all-PSCs, we monitored the photoluminescence (PL) quenching behaviors of the blend films as shown in Figure 6b-d. The neat $P_{\rm A}$ films and blend films were excited at 750 nm to maximize $P_{\rm A}$ absorption while minimizing $P_{\rm D}$ absorption. The calculated PL quenching efficiencies of the P(QxCN-T2) blend film exhibited the highest value of 87%. In comparison, the lower quenching efficiency (80%) was obtained in P(QxCN-TVT) blend film. In



contrast, the P(QxCN-T3) blend film showed a very low PL quenching efficiency of only 33%, suggesting poor charge-transfer. These trends are consistent with those observed in P(E,T) and corresponding J_{SC} values in the all-PSCs.

It is known that the PL guenching behaviors are also related to their blend morphologies, such as the $P_D - P_A$ interfacial area, at which exciton-dissociation occurs.^[24] Accordingly, the blend morphologies were investigated by atomic force microscopy (AFM). Figure S6 shows that the surface roughness of the P(QxCN-T3)-based blend film increased compared to those of P(QxCN-TVT)- and P(QxCN-T2)-based blend films as supported by its higher root-mean-square roughness (R_{a}) value. The P(QxCN-T2) blend film showed a relatively uniform and intermixed blend morphology, which can increase the $P_{\rm D}-P_{\rm A}$ interfacial area and promote efficient exciton-dissociation in all-PSCs during operation. Consequently, when considering the combined results of the electrical and morphological analyses, the efficient charge generation and transportation in the P(QxCN-T2) blend resulted in the enhanced PCE of all-PSCs with the improved J_{SC} and FF values.

Overall, we developed a new building block, QxCN, for ntype conjugated polymers and successfully demonstrated the potential of QxCN-based P_As for application in all-PSCs. To evaluate the current level of photovoltaic performance for QxCN-P_A-based all-PSCs, standard all-PSC devices using a wellknown P_A (N2200, poly[[N,N'-bis(2-octyldodecyl)-naphthalene-1,4,5,8-bis(dicarboximide)-2,6-diyl]-alt-5,5'-(2,2'-bithiophene)]) with the same PBDB-T $P_{\rm D}$ were fabricated and compared (Table S2).^[1f,4h,12d] The all-PSCs showed an average PCE of 6.56% with a $V_{\rm OC}$ of 0.89 V, a $J_{\rm SC}$ of 12.82 mA cm⁻², and a FF of 0.58. Thus, when compared to the standard device, the P(QxCN-T2)based device produced a higher $V_{\rm OC}$ value but lower $J_{\rm SC}$ and FF values. These results are mainly due to relatively low P(E,T) value (72%) and high S value (1.36 $k_{\rm B}Tq^{-1}$), which indicate that the photo-induced excitons do not effectively dissociate to free charges in this system. In addition, significant amount of the generated free charges become recombined before reaching the electrodes.^[12b,c,18a] The highest FF (0.49) of the P(QxCN-T2)based devices in this study is also still much lower than those of well-known high-performance all-PSCs. This encourages further optimization of the morphological properties of the QxCN-P_A based blend films in terms of domain size, purity, and polymer orientation. At the same time, insufficient electron affinity of QxCN-based P_A may hinder efficient exciton dissociation at the interfaces between the QxCN-based P_A and P_D . From the material-design perspective, we suspect that the presence of the electron-donating alkoxy side chains in the QxCN unit might reduce the n-type characteristics of the resulting $P_{\rm A}$ s.^[25] To address this issue, we are developing another series of QxCN-based P_{As} by (i) replacing the alkoxy-based side chains of QxCN with other appropriate side chains and (ii) copolymerizing QxCN unit with different counterparts that are more electrondeficient than T2 (e.g., 3,3'-difluoro-2,2'-bithiophene). Based on these design rules, we anticipate that the new QxCN-based P_{AS} will realize all-PSCs with enhanced photovoltaic performances.

Conclusions

In this work, we report the development of a series of Qxbased P_{AS} by introducing CN groups on the 6- and 7-position of Qx. In the series of QxCN-based P_As , their structural and electrical properties were investigated by modifying the electron-donating counterparts. The OFET measurements revealed that all the QxCN-based P_As had electron transport abilities with unipolar n-type characteristics. Among them, P(QxCN-T2) and P(QxCN-TVT) showed well-ordered semicrystalline structures, forming good vertical conducting pathways. As a result, P(QxCN-T2) and P(QxCN-TVT) demonstrated considerably high $\mu_{e,SCLC}$ of over 1.0×10^{-4} cm²V⁻¹s⁻¹. In the all-PSCs, the P(QxCN-T2)-based device achieved the highest PCE of 5.32% among the QxCN-based P_As, attributed to the higher electron mobility and exciton-dissociation probability with reduced charge recombination. Thus, we demonstrated that QxCN-based P_As can be applied as promising n-type components for organic electronics.

Experimental Section

Synthesis procedures for monomer and polymers

1,2-Bis(3,4-bis(octyloxy)phenyl)ethane-1,2-dione (1): 1,2-Bis (octyloxy)benzene^[9a] (7.0 g, 20.9 mmol) and AlCl₃ (1.4 g, 12.3 mmol) in 1,2-dichloroethane (DCE) were added to a solution of oxalyl chloride (1.6 g, 12.7 mmol) in DCE at 0 °C under nitrogen condition. After stirring for 30 min, the reaction solution was allowed to warm up to RT and further stirred overnight. The reaction mixture was poured to 1 M aqueous HCl solution and the separated organic layer was dried over MgSO₄. The crude product was purified by column chromatography (hexane/dichloromethane=1:1) to yield yellow powder. Yield: 57 %, ¹H NMR (400 MHz, CDCl₃): δ =7.55 (d, *J*=4 Hz, 2H), 7.44 (dd, *J*= 4 Hz, *J*=8 Hz, 2H), 6.85 (d, *J*=8 Hz, 2H), 4.04 (m, 8H), 1.84 (m, 8H), 1.46 (m, 8H), 1.28 (m, 32H), 0.88 ppm (m, 12H). MALDI-TOF MS: calculated for C₄₆H₇₄O₆ 722.55; found: 722.97 (*M*⁺).

4,5-Diamino-3,6-dibromophthalonitrile (2): 4,5-Diaminophthalonitrile (3.6 g, 22.8 mmol) was dried under vacuum and dissolved in degassed acetic acid under nitrogen condition. After cooling the solution to 0 °C, Br₂ (8.1 g, 50.5 mmol) was added dropwise and the reaction mixture was stirred overnight at RT. Aqueous Na₂S₂O₃ solution was poured into the mixture and precipitate was filtered to yield the product, which was used for the next reaction without further purification. Yield: 92%, ¹H NMR (400 MHz, DMSO-*d*₆): δ = 6.43 ppm (s, 4H). MALDI-TOF MS: calculated for C₈H₄Br₂N₄ 313.88; found: 314.48 (*M*⁺).

QxCN monomer (3): Compound 1 (3.7 g, 5.4 mmol) and 2 (1.7 g, 5.4 mmol) were dried under vacuum and dissolved in acetic acid under nitrogen condition. The reaction mixture was refluxed overnight and the solvent was condensed. The crude product was diluted with dichloromethane and washed with water. The organic phase was dried and purified by column chromatography (hexane/toluene=2:8) to provide orange powder. Yield: 61%, ¹H NMR (400 MHz, CDCl₃): δ =7.33 (s, 4H), 6.86 (d, *J*=12 Hz, 2H), 4.03 (m, 4H), 3.88 (m, 4H), 1.84 (m, 4H), 1.75 (m, 4H), 1.28 (m, 40H), 0.89 ppm (m, 12H). MALDI-TOF MS: calculated for C₅₄H₇₄Br₂N₄O₄ 1000.41; found: 1004.02 (*M*⁺).



Polymerization of QxCN-based $P_{A}s$: Compound 3 (1.0 equiv.), designated stannylated counterpart (5,5'-bis(trimethylstannyl)-2,2'-bithiophene, (*E*)-1,2-bis(5-(trimethylstannyl)thiophen-2-yl)ethene, or 5,5''-bis(trimethylstannyl)-2,2':5',2''-terthiophene) (1.0 equiv.), tris (dibenzylideneacetone)dipalladium(0) (Pd₂(dba)₃, 2 mol%), and tri(o-tolyl)phosphine (P(o-tol)₃, 8 mol%) were added to a reaction flask equipped with a magnetic stirrer. Dry toluene (5 mL) was injected and the resulting solution was bubbled with argon gas for 10 min. The reaction was heated at the condition of 120 °C for 2 h. After cooling to RT, the resulting solution was diluted with chloroform and precipitated in methanol. The precipitate was purified with Soxhlet extraction sequentially with methanol, acetone, hexane, dichloromethane, and chloroform. The polymers obtained from the chloroform fraction were precipitated in methanol again and dried in vacuum after filtration.

P(QxCN-T2): The target polymer was synthesized as per the general procedure described above using compound 3 (0.200 g, 0.199 mmol) and 5,5'-bis(trimethylstannyl)-2,2'-bithiophene (0.098 g, 0.199 mmol). Yield: 67 %, ¹H NMR (400 MHz, CDCl₃): δ = 8.27–7.80 (br, 2H), 7.73–7.30 (br, 4H), 7.02–6.68 (br, 4H), 4.38–3.40 (br, 8H), 2.07–0.48 ppm (br, 60H).

P(QxCN-TVT): The target polymer was synthesized as per the general procedure described above using compound 3 (0.200 g, 0.199 mmol) and (*E*)-1,2-bis(5-(trimethylstannyl)thiophen-2-yl) ethene (0.103 g, 0.199 mmol). Yield: 62 %, ¹H NMR (400 MHz, CDCl₃): δ = 8.20–7.77 (br, 2H), 7.71–7.33 (br, 4H), 7.02–6.49 (br, 6H), 4.41–3.47 (br, 8H), 2.08–0.40 ppm (br, 60H).

P(QxCN-T3): The target polymer was synthesized as per the general procedure described above using compound 3 (0.200 g, 0.199 mmol) and 5,5"-bis(trimethylstannyl)-2,2':5',2"-terthiophene (0.114 g, 0.199 mmol). Yield: 57%, ¹H NMR (400 MHz, CDCl₃): δ = 8.17–7.85 (br, 2H), 7.67–7.27 (br, 4H), 7.09–6.59 (br, 6H), 4.34–3.49 (br, 8H), 1.98–0.46 ppm (br, 60H).

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Conflict of Interest

The authors declare no conflict of interest.

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a) H. Benten, D. Mori, H. Ohkita, S. Ito, J. Mater. Chem. A 2016, 4, 5340;
 b) G. Wang, F. S. Melkonyan, A. Facchetti, T. J. Marks, Angew. Chem. Int. Ed. 2019, 58, 4129; Angew. Chem. 2019, 131, 4173; c) R. Zhao, J. Liu, L. Wang, Acc. Chem. Res. 2020, 53, 1557; d) J. Choi, W. Kim, S. Kim, T.-S. Kim, B. J. Kim, Chem. Mater. 2019, 31, 9057; e) Z. Genene, W. Mammo, E. Wang, M. R. Andersson, Adv. Mater. 2019, 31, 1807275; f) C. Lee, S. Lee, G. U. Kim, W. Lee, B. J. Kim, Macromolecules 2017, 50, 6861; h) N. B.

Kolhe, D. K. Tran, H. Lee, D. Kuzuhara, N. Yoshimoto, T. Koganezawa, S. A. Jenekhe, *ACS Energy Lett.* **2019**, *4*, 1162; i) Y. Zhang, Y. Xu, M. J. Ford, F. Li, J. Sun, X. Ling, Y. Wang, J. Gu, J. Yuan, W. Ma, *Adv. Energy Mater.* **2018**, *8*, 1800029; j) Y. Xu, J. Yuan, S. Zhou, M. Seifrid, L. Ying, B. Li, F. Huang, G. C. Bazan, W. Ma, *Adv. Funct. Mater.* **2019**, *29*, 1806747.

- [2] a) W. Wang, Q. Wu, R. Sun, J. Guo, Y. Wu, M. Shi, W. Yang, H. Li, J. Min, *Joule* 2020, *4*, 1070; b) Q. Fan, Q. An, Y. Lin, Y. Xia, Q. Li, M. Zhang, W. Su, W. Peng, C. Zhang, F. Liu, L. Hou, W. Zhu, D. Yu, M. Xiao, E. Moons, F. Zhang, T. D. Anthopoulos, O. Inganäs, E. Wang, *Energy Environ. Sci.* 2020, *13*, 5017; c) F. Peng, K. An, W. Zhong, Z. Li, L. Ying, N. Li, Z. Huang, C. Zhu, B. Fan, F. Huang, Y. Cao, *ACS Energy Lett.* 2020, *5*, 3702; d) H. Yu, Z. Qi, J. Yu, Y. Xiao, R. Sun, Z. Luo, A. M. H. Cheung, J. Zhang, H. Sun, W. Zhou, S. Chen, X. Guo, X. Lu, F. Gao, J. Min, H. Yan, *Adv. Energy Mater.* 2021, *11*, 2003171; e) H. Yao, F. Bai, H. Hu, L. Arunagiri, J. Zhang, Y. Chen, H. Yu, S. Chen, T. Liu, J. Y. L. Lai, Y. Zou, H. Ade, H. Yan, *ACS Energy Lett.* 2019, *4*, 417; f) Y. Li, Z. Jia, Q. Zhang, Z. Wu, H. Qin, J. Yang, S. Wen, H. Y. Woo, W. Ma, R. Yang, J. Yuan, *ACS Appl. Mater. Interfaces* 2020, *12*, 33028.
- [3] a) J. Choi, W. Kim, D. Kim, S. Kim, J. Chae, S. Q. Choi, F. S. Kim, T.-S. Kim, B. J. Kim, *Chem. Mater.* **2019**, *31*, 3163; b) N. Balar, J. J. Rech, R. Henry, L. Ye, H. Ade, W. You, B. T. O'Connor, *Chem. Mater.* **2019**, *31*, 5124; c) Q. Fan, W. Su, S. Chen, W. Kim, X. Chen, B. Lee, T. Liu, U. A. Méndez-Romero, R. Ma, T. Yang, W. Zhuang, Y. Li, Y. Li, T.-S. Kim, L. Hou, C. Yang, H. Yan, D. Yu, E. Wang, *Joule* **2020**, *4*, 658; d) J.-W. Lee, C. Sun, B. S. Ma, H. J. Kim, C. Wang, J. M. Ryu, C. Lim, T.-S. Kim, Y.-H. Kim, S.-K. Kwon, B. J. Kim, *Adv. Energy Mater.* **2021**, *11*, 2003367.
- [4] a) Z. Li, L. Ying, P. Zhu, W. Zhong, N. Li, F. Liu, F. Huang, Y. Cao, Energy Environ. Sci. 2019, 12, 157; b) L. Zhu, W. Zhong, C. Qiu, B. Lyu, Z. Zhou, M. Zhang, J. Song, J. Xu, J. Wang, J. Ali, W. Feng, Z. Shi, X. Gu, L. Ying, Y. Zhang, F. Liu, Adv. Mater. 2019, 31, 1902899; c) T. Kim, J.-H. Kim, T. E. Kang, C. Lee, H. Kang, M. Shin, C. Wang, B. Ma, U. Jeong, T.-S. Kim, B. J. Kim, Nat. Commun. 2015, 6, 8547; d) S. Chen, S. Jung, H. J. Cho, N.-H. Kim, S. Jung, J. Xu, J. Oh, Y. Cho, H. Kim, B. Lee, Y. An, C. Zhang, M. Xiao, H. Ki, Z.-G. Zhang, J.-Y. Kim, Y. Li, H. Park, C. Yang, Angew. Chem. Int. Ed. 2018, 57, 13277; Angew. Chem. 2018, 130, 13461; e) M. Kim, H. I. Kim, S. U. Ryu, S. Y. Son, S. A. Park, N. Khan, W. S. Shin, C. E. Song, T. Park, Chem. Mater. 2019, 31, 5047; f) B. Lin, L. Zhang, H. Zhao, X. Xu, K. Zhou, S. Zhang, L. Gou, B. Fan, L. Zhang, H. Yan, X. Gu, L. Ying, F. Huang, Y. Cao, W. Ma, Nano Energy 2019, 59, 277; g) J.-W. Lee, B. S. Ma, J. Choi, J. Lee, S. Lee, K. Liao, W. Lee, T.-S. Kim, B. J. Kim, Chem. Mater. 2020, 32, 582; h) N. Zhou, A. Facchetti, Mater. Today 2018, 21, 337; i) R. Matsidik, H. Komber, A. Luzio, M. Caironi, M. Sommer, J. Am. Chem. Soc. 2015, 137, 6705; j) R. Tkachov, Y. Karpov, V. Senkovskyy, I. Raguzin, J. Zessin, A. Lederer, M. Stamm, B. Voit, T. Beryozkina, V. Bakulev, W. Zhao, A. Facchetti, A. Kiriy, Macromolecules 2014, 47, 3845.
- [5] a) S. Shi, P. Chen, Y. Chen, K. Feng, B. Liu, J. Chen, Q. Liao, B. Tu, J. Luo, M. Su, H. Guo, M. G. Kim, A. Facchetti, X. Guo, Adv. Mater. 2019, 31, 1905161; b) Z.-G. Zhang, Y. Yang, J. Yao, L. Xue, S. Chen, X. Li, W. Morrison, C. Yang, Y. Li, Angew. Chem. Int. Ed. 2017, 56, 13503; Angew. Chem. 2017, 129, 13688; c) H. Kang, W. Lee, J. Oh, T. Kim, C. Lee, B. J. Kim, Acc. Chem. Res. 2016, 49, 2424.
- [6] a) R. Zhao, N. Wang, Y. Yu, J. Liu, Chem. Mater. 2020, 32, 1308; b) K. Feng, J. Huang, X. Zhang, Z. Wu, S. Shi, L. Thomsen, Y. Tian, H. Y. Woo, C. R. McNeill, X. Guo, Adv. Mater. 2020, 32, 2001476; c) X. Long, Z. Ding, C. Dou, J. Zhang, J. Liu, L. Wang, Adv. Mater. 2016, 28, 6504; d) Y. Shi, H. Guo, J. Huang, X. Zhang, Z. Wu, K. Yang, Y. Zhang, K. Feng, H. Y. Woo, R. P. Ortiz, M. Zhou, X. Guo, Angew. Chem. Int. Ed. 2020, 59, 14449; Angew. Chem. 2020, 132, 14557.
- [7] a) J. Yuan, J. Ouyang, V. Cimrová, M. Leclerc, A. Najari, Y. Zou, J. Mater. Chem. C 2017, 5, 1858; b) D. Gedefaw, M. Prosa, M. Bolognesi, M. Seri, M. R. Andersson, Adv. Energy Mater. 2017, 7, 1700575.
- [8] a) C. Sun, F. Pan, H. Bin, J. Zhang, L. Xue, B. Qiu, Z. Wei, Z. G. Zhang, Y. Li, *Nat. Commun.* **2018**, *9*, 743; b) Y. Li, M. Kim, Z. Wu, C. Lee, Y. W. Lee, J.-W. Lee, Y. J. Lee, E. Wang, B. J. Kim, H. Y. Woo, *J. Mater. Chem. C* **2019**, *7*, 1681; c) H.-C. Chen, Y.-H. Chen, C.-C. Liu, Y.-C. Chien, S.-W. Chou, P.-T. Chou, *Chem. Mater.* **2012**, *24*, 4766; d) E. Wang, L. Hou, Z. Wang, S. Hellström, F. Zhang, O. Inganäs, M. R. Andersson, *Adv. Mater.* **2010**, *22*, 5240.
- [9] a) Y. Li, S.-J. Ko, S. Y. Park, H. Choi, T. L. Nguyen, M. A. Uddin, T. Kim, S. Hwang, J. Y. Kim, H. Y. Woo, *J. Mater. Chem. A* **2016**, *4*, 9967; b) D. Liu, W. Zhao, S. Zhang, L. Ye, Z. Zheng, Y. Cui, Y. Chen, J. Hou, *Macromolecules* **2015**, *48*, 5172; c) Y. Lu, Z. Xiao, Y. Yuan, H. Wu, Z. An, Y. Hou, C. Gao, J. Huang, *J. Mater. Chem. C* **2013**, *1*, 630.
- [10] a) A. Casey, S. D. Dimitrov, P. Shakya-Tuladhar, Z. Fei, M. Nguyen, Y. Han, T. D. Anthopoulos, J. R. Durrant, M. Heeney, *Chem. Mater.* 2016, 28,



5110; b) H. J. Yun, S. J. Kang, Y. Xu, S. O. Kim, Y. H. Kim, Y. Y. Noh, S. K. Kwon, *Adv. Mater.* **2014**, *26*, 7300.

- [11] a) J. Wudarczyk, G. Papamokos, V. Margaritis, D. Schollmeyer, F. Hinkel, M. Baumgarten, G. Floudas, K. Mîllen, *Angew. Chem. Int. Ed.* **2016**, *55*, 3220; *Angew. Chem.* **2016**, *128*, 3275; b) H.-H. Cho, S. Kim, T. Kim, V. G. Sree, S.-H. Jin, F. S. Kim, B. J. Kim, *Adv. Energy Mater.* **2018**, *8*, 1701436.
- [12] a) S. Feng, C. Liu, X. Xu, X. Liu, L. Zhang, Y. Nian, Y. Cao, J. Chen, ACS Macro Lett. 2017, 6, 1310; b) N. B. Kolhe, H. Lee, D. Kuzuhara, N. Yoshimoto, T. Koganezawa, S. A. Jenekhe, Chem. Mater. 2018, 30, 6540; c) J.-W. Lee, M. J. Sung, D. Kim, S. Lee, H. You, F. S. Kim, Y.-H. Kim, B. J. Kim, S.-K. Kwon, Chem. Mater. 2020, 32, 2572; d) S. Seo, J. Kim, H. Kang, J.-W. Lee, S. Lee, G.-U. Kim, B. J. Kim, Macromolecules 2021, 54, 53.
- [13] J. Choi, K.-H. Kim, H. Yu, C. Lee, H. Kang, I. Song, Y. Kim, J. H. Oh, B. J. Kim, Chem. Mater. 2015, 27, 5230.
- [14] M. M. Szumilo, E. H. Gann, C. R. McNeill, V. Lemaur, Y. Oliver, L. Thomsen, Y. Vaynzof, M. Sommer, H. Sirringhaus, *Chem. Mater.* 2014, 26, 6796.
- [15] a) L. Wang, J. S. Park, H. G. Lee, G.-U. Kim, D. Kim, C. Kim, S. Lee, F. S. Kim, B. J. Kim, ACS Appl. Mater. Interfaces 2020, 12, 56240; b) H. Chen, Y. Guo, Z. Mao, G. Yu, J. Huang, Y. Zhao, Y. Liu, Chem. Mater. 2013, 25, 3589.
- [16] a) R. Zhao, C. Dou, Z. Xie, J. Liu, L. Wang, *Angew. Chem. Int. Ed.* 2016, *55*, 5313; *Angew. Chem.* 2016, *128*, 5399; b) T. L. Nguyen, H. Choi, S. J. Ko, M. A. Uddin, B. Walker, S. Yum, J. E. Jeong, M. H. Yun, T. J. Shin, S. Hwang, J. Y. Kim, H. Y. Woo, *Energy Environ. Sci.* 2014, *7*, 3040.
- [17] B. Capozzi, E. J. Dell, T. C. Berkelbach, D. R. Reichman, L. Venkataraman, L. M. Campos, J. Am. Chem. Soc. 2014, 136, 10486.
- [18] a) B. Fan, L. Ying, Z. Wang, B. He, X.-F. Jiang, F. Huang, Y. Cao, *Energy Environ. Sci.* 2017, *10*, 1243; b) L. Gao, Z. G. Zhang, L. Xue, J. Min, J. Zhang, Z. Wei, Y. Li, *Adv. Mater.* 2016, *28*, 1884; c) C. Lee, H. Kang, W. Lee, T. Kim, K. H. Kim, H. Y. Woo, C. Wang, B. J. Kim, *Adv. Mater.* 2015, *27*, 2466.

- [19] Z. Wu, C. Sun, S. Dong, X.-F. Jiang, S. Wu, H. Wu, H.-L. Yip, F. Huang, Y. Cao, J. Am. Chem. Soc. 2016, 138, 2004.
- [20] a) S. W. Kim, Y. Wang, H. You, W. Lee, T. Michinobu, B. J. Kim, ACS Appl. Mater. Interfaces 2019, 11, 35896; b) H. You, D. Kim, H.-H. Cho, C. Lee, S. Chong, N. Y. Ahn, M. Seo, J. Kim, F. S. Kim, B. J. Kim, Adv. Funct. Mater. 2018, 28, 1803613; c) J. Jung, W. Lee, C. Lee, H. Ahn, B. J. Kim, Adv. Energy Mater. 2016, 6, 1600504.
- [21] S. R. Cowan, A. Roy, A. J. Heeger, Phys. Rev. B 2010, 82, 245207.
- [22] a) C. M. Proctor, J. A. Love, T. Q. Nguyen, *Adv. Mater.* 2014, *26*, 5957;
 b) D. Bartesaghi, I. d. C. Pérez, J. Kniepert, S. Roland, M. Turbiez, D. Neher, L. J. A. Koster, *Nat. Commun.* 2015, *6*, 7083.
- [23] a) K. Xian, Y. Cui, Y. Xu, T. Zhang, L. Hong, H. Yao, C. An, J. Hou, J. Phys. Chem. C 2020, 124, 7691; b) J. Miao, H. Li, T. Wang, Y. Han, J. Liu, L. Wang, J. Mater. Chem. A 2020, 8, 20998.
- [24] a) J. A. Bartelt, Z. M. Beiley, E. T. Hoke, W. R. Mateker, J. D. Douglas, B. A. Collins, J. R. Tumbleston, K. R. Graham, A. Amassian, H. Ade, J. M. J. Fréchet, M. F. Toney, M. D. McGehee, *Adv. Energy Mater.* 2013, *3*, 364; b) C. Lee, Y. Li, W. Lee, Y. Lee, J. Choi, T. Kim, C. Wang, E. D. Gomez, H. Y. Woo, B. J. Kim, *Macromolecules* 2016, *49*, 5051; c) Z. Li, X. Xu, W. Zhang, X. Meng, W. Ma, A. Yartsev, O. Inganäs, M. R. Andersson, R. A. J. Janssen, E. Wang, *J. Am. Chem. Soc.* 2016, *138*, 10935.
- [25] a) Y. Huang, M. Zhang, L. Ye, X. Guo, C. C. Han, Y. Li, J. Hou, J. Mater. Chem. 2012, 22, 5700; b) N. J. Jeon, H. G. Lee, Y. C. Kim, J. Seo, J. H. Noh, J. Lee, S. I. Seok, J. Am. Chem. Soc. 2014, 136, 7837.

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FULL PAPERS

The development of cyano-func-

tionalized quinoxaline (QxCN)-based polymer acceptors (P_A s) is reported. The resulting P_{A} s exhibit significant electron mobilities of over $10^{-2} \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$ in organic transistors and a power conversion efficiency of 5.3% in all-polymer solar cells.

Quinoxaline-Based n-type Polymers



H. You, Dr. H. Kang, D. Kim, J. S. Park, J.-W. Lee, S. Lee, Prof. F. S. Kim, Prof. B. J. Kim*



P(QxCN-T2) PCE: 5.3 %

Cyano-Functionalized Quinoxaline-**Based Polymer Acceptors for All-**Polymer Solar Cells and Organic Transistors

