1. Introduction

Currently, polymer solar cells (PSCs) with bulk-heterojunction (BHJ) composite layers have attracted broad attention due to their potential applications for large area, low cost, and flexible renewable power sources.\(^1\) Although the power conversion efficiency (PCE) of PSCs was already promoted to \(\sim 10\%\)\(^1\) compared with silicon solar cells or other thin-film counterparts, high PCE for PSCs was pursued for its commercialization.\(^2\)–\(^8\) One key factor to obtain desired photovoltaic properties of PSCs is the development of promising donor conjugated polymers with a broad absorption range and a low-lying highest occupied molecular orbital (HOMO) energy level of the solar cells had been optimized.\(^9\)–\(^11\) One effective approach is to introduce a strong electron withdrawing moiety and the HOMO and band gaps of the quinoxaline-based alternating polymers could be effectively modulated due to the tunable structure of quinoxaline unit.\(^12\)–\(^20\) Therefore, fluorinated quinoxaline-based alternating polymers showed low-lying HOMO and improved photovoltaic performance.\(^21\)–\(^31\) Herein, a new alternating polymer poly([N-9'-heptadecyl-2,7-carbazole]-alt-5,5-[5',8'-di-2-thienyl-(6'-fluoro-2',3'-bis-(3'-octyloxyphenyl)-quinoxaline)]) ([PCzFTQx]) based on mono-fluorinated quinoxaline and 2,7-carbazole derivative was synthesized. Benefit from its lowered HOMO energy level (\(-5.31\) eV) and PCE (5.19\%) of \(0.94\) V, relatively to those of 4.72\% and 0.82\% for PCzTQx-based device. The experimental data indicated that fluorinated quinoxaline based polymer [PCzFTQx] should be a promising donor for polymer solar cells.
2. Experimental

2.1 Materials and characterization

All chemicals and solvents were purchased from Aldrich or Alfa & Aesar. Tetrahydrofuran (THF) is dried over Na/benzophenone ketyl and freshly distilled prior to use. 5,8-Bis(5-bromothiophen-2-yl)-2,3-bis(3-octyloxy)phenyl)quinoxaline (M1) and 5,8-bis(5-bromothiophen-2-yl)-6-fluoro-2,3-bis(3-octyloxy)phenyl)quinoxaline (M2) that followed our previous work7 were prepared. 2,7-Bis(4’4’,5’,5’,6-tetramethyl-1’,3’,2’-dioxaborolan-2’-yl)-N9’-heptadecancarbazole (M3) were synthesized according to the reported literatures. The synthetic routes of monomers and copolymers are given in Scheme 1.

2.2 Synthesis of PCzFTQx and PCzTQx

In a 50 mL dry flask, monomer M1 (175.7 mg, 0.2 mmol) or M2 (172.2 mg, 0.2 mmol) and 2,7-bis(4’,4’,5’,5’,6-tetramethyl-1’,3’,2’-dioxaborolan-2’-yl)-N9’-heptadecanecarbazole M3 (131.5 mg, 0.2 mmol) were dissolved in a mixture of 20% aqueous tetrahydrofuran (THF) solution. Polymers in chloroform solution (analytical reagent, 2 mg mL\(^{-1}\)) at the end of polymerization. A methylthiophosphate (PVP, AI 4083, H. C. Starck Inc.) layer was spin-coated from an 1:3 (the optimal specimen in the previous literature23) was used as the anode. The ITO glass was ultrasonically cleaned with detergent, distilled water, acetone, isopropyl alcohol, and then treated with ultraviolet-ozone (Ming Heng, PDC-MG) for 20 min. Then a 40 nm thick PEDOT: PSS (Clevios P VP Al 4083, H. C. Starck Inc.) layer was spin-coated from an aqueous solution onto ITO substrates, followed by being annealed at 120 °C for 30 min on a hotplate in air. The substrates were transferred into a nitrogen-filled glove box. Then the photoactive layers were prepared by spin-coating

2.3 Characterization

All the compounds were characterized by nuclear magnetic resonance spectra (NMR) recorded (Bruker AV 500 spectrometer) in chloroform-d (CDCl\(_3\)) at room temperature using tetramethylsilane (TMS) as an internal reference. The chemical shifts were accounted in ppm related to the singlet of CDCl\(_3\) at 7.26 ppm for \(^1\)H NMR. Molecular weights and distributions of the copolymers were estimated by gel permeation chromatography (GPC) method, tetrahydrofuran (THF) as eluent and polystyrene as standard. The absorption spectra and thermogravimetric analysis (TGA, as shown in Fig. S2 in the ESI†) were measured with a Unico UV-2102 scanning spectrophotometer and a Universal V2.6D TA instrument, respectively. The electrochemical cyclic voltammetry (CV) was determined by a CHI 660D Electrochemical Workstation with glassy carbon, Pt wire, and Ag/Ag\(^{+}\) electrodes as the working electrode, counter electrode, and reference electrode respectively in a 0.1 mol L\(^{-1}\) tetrabutylammonium hexafluorophosphate (Bu\(_4\)NPF\(_6\)) acetonitrile solution. Polymers in chloroform solution (analytical reagent, 2 mg mL\(^{-1}\)) were drop casted onto working electrode and dried in air at room temperature. Atomic force microscopy (AFM) measurement of the surface morphology of samples was conducted in air under ambient condition using the PicoPlus microscope (Agilent).

2.4 PSCs fabrication and characterization

PSCs with the configuration of ITO/PEDOT : PSS/PCzFTQx : PCzTQx : PC\(_{71}\)BM/poly[(9,9-bis(3’-(N,N-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctyfluorene)] (PFN)/Al were fabricated and characterized in this work. With varying the weight ratio of PCzFTQx : PC\(_{71}\)BM from 1 : 1 to 1 : 4, the photovoltaic properties of PSCs were systematically investigated. For comparison, the control device based on PCzTQx : PC\(_{71}\)BM (1 : 3) (the optimal specimen in the previous literature23) was also prepared with the same configuration and experimental condition in this work.

ITO-coated glass with sheet resistance of ~15 Ω per square (\(\Omega \square^{-1}\)) was used as the anode. The ITO glass was ultrasonically cleaned with detergent, distilled water, acetone, isopropyl alcohol, and then treated with ultraviolet-ozone (Ming Heng, PDC-MG) for 20 min. Then a 40 nm thick PEDOT : PSS (Clevios P VP Al 4083, H. C. Starck Inc.) layer was spin-coated from an aqueous solution onto ITO substrates, followed by being annealed at 120 °C for 30 min on a hotplate in air. The substrates were transferred into a nitrogen-filled glove box. Then the photoactive layers were prepared by spin-coating

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**Scheme 1** Synthetic route of PCzFTQx and PCzTQx.
(1000 rpm, 30 s) a blend solution of the polymer and PC_{71}BM in ortho-dichlorobenzene (oDCB) on the top of ITO/PEDOT : PSS substrates. The thickness of the photoactive layers was controlled in the range of 80–100 nm. A 5 nm PFN layer was then spin-coated from methanol solution in the presence of a trace amount of acetic acid onto the active layer. The thin PFN layer was employed as the cathode interlayer since it could improve the electron extraction. Finally, the substrates were transferred into a vacuum chamber and Al cathodes (~120 nm) were thermally deposited onto the substrates. The effective area of PSCs was ~0.16 cm² controlled by the shadow mask. The photovoltaic properties of PSCs were recorded by J–V measurement (using a Keithley 2400 source meter) under 1 sun, AM 1.5G spectrum from a solar simulator (Newport model 94021A, 100 mW cm⁻²). A monocrystal silicon cell (VLSI Standards Inc.) calibrated by the National Renewable Energy Laboratory (NREL) is used as a reference. The incident photo to current conversion efficiency spectra were measured by using a Hypermonolight System (QTEST 1000 AD, Crowntech Inc.).

### 3. Results and discussion

#### 3.1 Thermal properties

As shown in the TGA curves of the polymers (Fig. S2 in the ESI†), the thermal decomposition temperature (5% weight loss) of PCzFTQx was about 403°C, slightly lower than that of PCzTQx (405°C), indicating its good thermal stability for photovoltaic applications.

#### 3.2 Absorption properties

The normalized UV-vis absorption spectra of PCzFTQx and PCzTQx in chloroform (CHCl₃) solution were exhibited in Fig. 1. The absorption peaks of PCzFTQx and PCzTQx in CHCl₃ solution were almost identical. The peaks located at 386 nm and 517 nm, which were attributed to the absorption of carbazole segment and intramolecular charge transfer, respectively. As to the film state, the absorption peaks of PCzFTQx and PCzTQx in the long wavelength region were red shifted to 536 nm and 542 nm, respectively. The weak bathochromic shift effect of the polymers from solution state to film state implied that the aggregation and chain–chain stacking of PCzFTQx and PCzTQx were relatively limited. The optical band gap ($E_{\text{opt}}$) of the polymers

### Table 1: Electrochemical potentials and energy levels of PCzFTQx and PCzTQx

| Polymers    | $E_{\text{ox}}^{\text{onset}}$ (V) | $E_{\text{red}}^{\text{onset}}$ (V) | $E_{\text{HOMO}}$ (eV) | $E_{\text{LUMO}}$ (eV) | $E_{\text{HOMO}}^{\text{opt}}$ (eV) | $E_{\text{LUMO}}^{\text{opt}}$ (eV) |
|------------|----------------------------------|------------------------------------|------------------------|------------------------|-----------------------------------|-----------------------------------|
| PCzFTQx    | 0.59                             | -1.62                              | -5.31                  | -3.10                  | 2.21                              | 2.04                              |
| PCzTQx     | 0.50                             | -1.64                              | -5.22                  | -3.08                  | 2.14                              | 1.98                              |
was calculated to be 2.04 eV for PCzFTQx and 1.98 eV for PCzTQx from the absorption onset of the films.

3.3 Electrochemical properties

In order to study the electrochemical properties of the polymers, cyclic voltammetry (CV) was carried out using an electrochemical workstation. CV curves of PCzFTQx and PCzTQx films were measured as shown in Fig. 2. The level of ferrocene/ferrocinium (Fc/Fc⁺) was −4.72 eV below the vacuum level.²⁹ The former potential of Fc/Fc⁺ was ~0.08 eV against Ag/Ag⁺. The onset oxidation and reduction potential of PCzFTQx film located at 0.59 V and −1.62 V, respectively. Therefore, the

Fig. 4 Molecular structures and electron density of LUMO+1, LUMO, HOMO and HOMO−1 for PCzTQx and PCzFTQx computed at the B3LYP/6-31G* level.
HOMO and LUMO of PCzFTQx were evaluated to be $-5.31 \text{ eV}$ and $-3.08 \text{ eV}$, respectively. The electrochemical energy band gap ($\Delta E_{\text{opt}}$) of PCzFTQx film was calculated to be $2.21 \text{ eV}$. Although $\Delta E_{\text{opt}}$ value was a little bit larger than $\Delta E_{\text{opt}}$ (2.04 eV) estimated from the UV-vis spectra, it was still in the range of error (0.1–0.5 eV). As to PCzTQx film, the HOMO and LUMO were measured to be $-5.22 \text{ eV}$ and $-3.08 \text{ eV}$, respectively. The $\Delta E_{\text{opt}}$ and $\Delta E_{\text{opt}}$ of PCzTQx were $2.14 \text{ eV}$ and $1.98 \text{ eV}$, slightly smaller than those of PCzFTQx. The energy level data were summarized in Table 1 and the schematic illustration of energy levels of PCzFTQx, PCzTQx, PC$_{71}$BM$_{14}$ and PEDOT : PSS was depicted in Fig. 3. The HOMO of PCzFTQx was $-0.09 \text{ eV}$ deeper than that of PCzTQx, which was ascribed to the introduction of fluorine atom onto the quinoxaline segment in PCzFTQx. Therefore, the gap between the HOMO level of PCzFTQx and LUMO level of PC$_{71}$BM was as large as $1.40 \text{ eV}$, which facilitated high $V_{\text{oc}}$ of photovoltaic devices. Moreover, the LUMO level of PCzFTQx was $-0.81 \text{ eV}$ higher than that of PC$_{71}$BM, then electrons could transfer automatically at the donor/acceptor interface.

### 3.4 Theoretical calculations

Quantum chemistry calculation by the DFT (B3LYP/6-31G* level) method was employed to demonstrate the electronic structures of the PCzTQx and PCzFTQx. Three repeat units were chosen as the model compounds for the simulations of PCzTQx and PCzFTQx, and the actual long alkyl groups were kept without simplification. As shown in Fig. 4, the electron density in HOMO orbitals distributed along the whole backbone despite somewhat more intensely at carbazole conjugated units and less at quinoxaline units. However, the electron density in LUMO orbitals mainly localized at the quinoxaline units. The HOMO–1 electron density distributed more intensely in π-bridge thiophene and carbazole conjugated units, while the LUMO+1 electron density distributed more intensively at the quinoxaline units. The electron density distribution of the different orbitals implied that the internal charge transfers were possible in the conjugated systems. It should be noted that the F atom that incorporated onto the quinoxaline unit showed significant effect on the HOMO and LUMO energy levels of PCzFTQx ($-4.64 \text{ eV}$/$-2.26 \text{ eV}$). The additional F atoms depressed both HOMO and LUMO energy levels, which agreed well with the trend of electrochemical measurements.

### 3.5 Photovoltaic properties

In order to study the photovoltaic properties of PCzFTQx, PSCs with the conventional structure of ITO/PEDOT : PSS/PCzFTQx : PC$_{71}$BM/PFN/Al were fabricated. PCzFTQx was used as the electron donor and PC$_{71}$BM as the electron acceptor. Chlorobenzene was used as the solvent to prepare the PCzFTQx : PC$_{71}$BM blend films. The weight ratio of PCzFTQx : PC$_{71}$BM was changed from 1 : 1 to 1 : 4 and $J$–$V$ curves of PSCs were modulated accordingly as shown in Fig. 5. When the weight ratio of PCzFTQx : PC$_{71}$BM reached 1 : 3, the PSC exhibited a reasonable photovoltaic performance as shown in Table 2 and the $V_{\text{oc}}, J_{\text{sc}}$, FF and PCE values were evaluated to be $0.94 \text{ V}$, $9.71 \text{ mA cm}^{-2}$, 0.56 and 5.09%, respectively. As expected, the PSC based on PCzFTQx : PC$_{71}$BM (1 : 3) has higher $V_{\text{oc}}$ and PCE than those of the corresponding PCzTQx-based device (blend ratio of 1 : 3), which was ascribed to the introduction of fluorine atom into the polymer and the lowering HOMO level of PCzFTQx. It was worthy to note that the PSC with PCzFTQx : PC$_{71}$BM (1 : 3) as the active layer exhibited the highest $J_{\text{sc}}$ of 9.71 mA cm$^{-2}$ in this work, approximately 0.14 mA cm$^{-2}$ higher than that of PCzTQx control device. Moreover, when the weight ratio of PCzFTQx : PC$_{71}$BM reached 1 : 4, the highest PCE of 5.19% was achieved, mainly due to the increased FF of 0.59. Therefore, the PCE value was almost unaltered when the ratio of PCzFTQx : PC$_{71}$BM changed from 1 : 3 to 1 : 4.

The external quantum efficiency (EQE) curves were also recorded to characterize the photovoltaic properties of the PSCs as shown in Fig. 6. The EQE curves covered from 350 nm to 650 nm, which matched well with the absorption range of the organic photoactive layers. Compared with any other PCzFTQx : PC$_{71}$BM-based devices with different blend ratios, the PCzFTQx : PC$_{71}$BM (1 : 3)-based device exhibited a high EQE value up to 62%, leading to a high short circuit current density of 9.71 mA cm$^{-2}$.

Atomic force microscopy (AFM) was conducted to evaluate the phase-separated morphologies of the blend films and the topography images (5 μm × 5 μm) of PCzTQx : PC$_{71}$BM (1 : 3) and PCzFTQx : PC$_{71}$BM (1 : 4) corresponding to the best photovoltaic performance were shown in Fig. 7. The
interpenetrating feature with bicontinuous network between the polymer and PC$_{71}$BM was realized for both the two blend films. PCzTQx : PC$_{71}$BM blend film showed a relatively smooth surface with a root-mean-square roughness (RMS) of 2.99 nm and an ideal domain size of 10–20 nm, which were favorable for efficient diffusion of excitons to donor–acceptor interfaces. However, for the fluorinated polymer PCzFTQx-based blend film, RMS value was enlarged (5.27 nm) with the corresponding domain size of 40–60 nm, which was somewhat larger than the typical exciton diffusion length of ca. 10 nm, thus was not desired for efficient exciton dissociation at the D/A interfaces.

To get insight into the charge transporting property of the two polymers, the hole-only devices were fabricated to measure the hole mobility using the space-charge-limited current (SCLC) method. The hole-only devices employed the structure of ITO/PEDOT : PSS/polymer : PC$_{71}$BM/MoO$_3$/Al. And the $J$–$V$ characteristics of the hole-only devices were measured and fit the dark current using the SCLC model, which was described by the equation: $J = \frac{9}{8} \varepsilon_0 \mu \left( \frac{V_{\text{app}} - V_s - V_{\text{bi}}} {L^2} \right)$, where $J$ was the current density, $\varepsilon_0$ was the permittivity of free space, $\mu$ was the charge carrier mobility, $V_{\text{app}}$ was the applied voltage, $V_s$ was the voltage drop from the substrate’s series resistance and $V_{\text{bi}}$ was the built-in voltage. As illustrated in Fig. S3, the hole ($\mu_h$) mobilities were calculated through the slope of the $J^{1/2}$–$V$ curves, which were exhibited in Table S1. Relative to the $\mu_h$ value of 3.12 × 10$^{-5}$ cm$^2$ V$^{-1}$ s$^{-1}$ for PCzTQx, the fluorinated polymer PCzFTQx showed slightly high $\mu_h$ value of 6.18 × 10$^{-5}$ cm$^2$ V$^{-1}$ s$^{-1}$, which was consistent with the experimental data of the previous literature. And the high $\mu_h$ of PCzFTQx was beneficial to the photovoltaic performance of the corresponding devices.

The absorption spectra of PCzTQx : PC$_{71}$BM and PCzFTQx : PC$_{71}$BM active layers under the optimal weight ratios were recorded in Fig. 8. It can be seen that in the region from 300 nm to 440 nm, the PCzFTQx blend film showed slightly high absorption property, which was mainly resulted from the high ratio of PC$_{71}$BM in the blend film. In addition, a little stronger absorption for PCzFTQx blend film in the range of 440–600 nm might be ascribed to the slightly enhanced absorption behavior of PCzFTQx pristine film in this region as

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**Fig. 6** EQE spectra of the PSCs.

**Fig. 7** AFM topography images (5 μm × 5 μm) of ITO/PEDOT : PSS/PCzTQx : PC$_{71}$BM (1 : 3) (left, RMS = 2.99 nm), and ITO/PEDOT : PSS/PCzFTQx : PC$_{71}$BM (1 : 4) (right, RMS = 5.27 nm).

**Fig. 8** Absorption spectra of PCzTQx : PC$_{71}$BM (1 : 3) and PCzFTQx : PC$_{71}$BM (1 : 4) films.
seen in Fig. 1, which would be benefit to the improvement of photocurrent. As described ahead, the domain size of PCzFTQx:PC71BM blend film was larger to some extent and not suitable for the efficient exciton dissociation. Then, the $J_{sc}$ of the corresponding PCzFTQx:PC71BM based device was 9.31 mA cm$^{-2}$, almost equal to that of PCzTQx:PC71BM based device. Therefore, if further improvement of morphology can be adopted to obtain ideal domain size and phase separation, enhanced photovoltaic performance may be anticipated for PCzFTQx based solar cells.

4. Conclusions

A novel copolymer PCzFTQx with mono-fluorinated quinoxaline derivative and 2,7-carbazole segment was reported and the photovoltaic properties of PCzFTQx were investigated in this work. Due to the introduction of fluorine atom in quinoxaline unit, PCzFTQx exhibited a lowered HOMO level (−5.31 eV), about 0.09 eV deeper than that of the non-fluorinated counterpart PCzTQx (−5.22 eV). High $V_{oc}$ (0.94 V) and PCE (5.19%) were achieved for the optimized PCzFTQx:PC71BM-based PSCs, which were 12% and 10% larger than those of the non-fluorinated analogue PCzTQx-based devices. Therefore, the fluorine atom in PCzFTQx was an effective electron-withdrawing unit and the accompanying beneficial modulation of the energy levels for PCzFTQx was realized accordingly. Hence, PCzFTQx showed remarkable photovoltaic properties and would be a promising donor candidate for PSCs and other optoelectronic devices.

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