

Supporting Information

Complementary Semiconducting Polymer Blends: the Influence of Side Chains of Matrix Polymers

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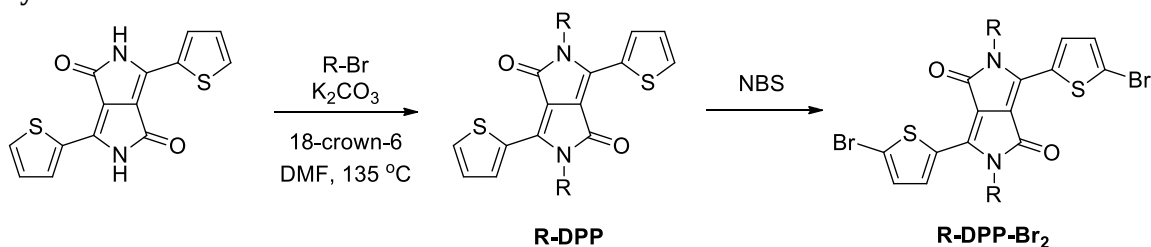
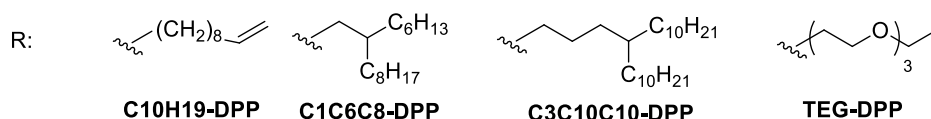
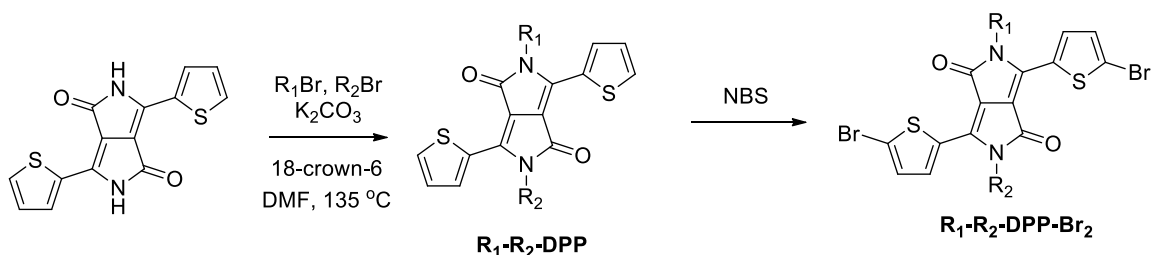
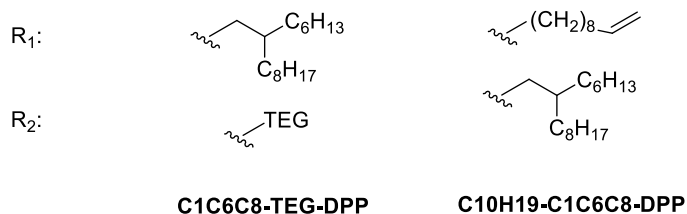
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Experiment Section

All reagents were purchased from Sigma-Aldrich and used without further purification. 1,4-diketo-3,6-di(thiophene-2-yl)pyrrolo[3,4-*c*]pyrrole (**DPP**)¹, 3,6-bis(5-bromothiophen-2-yl)-2,5-bis(2-hexyldecyl)pyrrolo[3,4-*c*]pyrrole-1,4(*2H,5H*)-dione (**C1C6C8-DPP-Br₂**)² and 3,6-bis(5-bromothiophen-2-yl)-2,5-bis(4-decyltetradecyl)pyrrolo[3,4-*c*]pyrrole-1,4(*2H,5H*)-dione (**C3C10C10-DPP-Br₂**)³ were synthesized according to the methods previously reported, respectively.

Synthesis of 1-Bromo-2-(2-(2-ethoxyethoxy)ethoxy)ethane (TEG-Br): Tri(ethylene glycol) monoethyl ether (10.0 g, 56.1 mmol) and triphenylphosphine (9.5 g, 53 mmol) were dissolved in 50 mL of dichloromethane. The solution was cooled to 0 °C with ice water bath. *N*-bromosuccinimide (14.0 g, 53.3 mmol) was added to the solution in small fractions. The reaction was kept overnight. Solvent was removed under vacuum and the mixture was taken up in hexanes and filtered. Product was concentrated by removal and hexanes and purified using flash column chromatography using hexanes-dichloromethane (3:1) as the eluent. Product was obtained as a colourless oil (9.53 g, 39.5 mmol, 70.4%). ¹H NMR (400 MHz, CDCl₃) δ 3.81 (t, *J* = 6.4 Hz, 2H), 3.69 – 3.46 (m, 12H), 1.21 (t, *J* = 7.0 Hz, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 71.11, 70.65, 70.51, 70.45, 69.72, 66.55, 30.19, 15.06. MS (ESI, M+Na⁺) for C₈H₁₇BrNaO₃: *calc'd* 263.0253 *found* 263.0257.

Synthesis of the monomers:



General method for the synthesis of DPP derivatives: DPP (1.53 g, 5.1 mmol), potassium carbonate (2.11 g, 15.3 mmol) and 18-crown-6 (60 mg, 0.23 mmol) were dissolved in anhydrous *N,N*-dimethylformamide (30 mL) in an oven dried flask. The solution was stirred at 100 °C for 1 hour under nitrogen. The corresponding alkyl bromide (11.2 mmol) was added dropwise via a syringe (For the asymmetric DPP derivatives, a 1:1 mixture of the corresponding alkyl bromides was added). The solution was left stirring at 100 °C overnight. After cooling down to room temperature, 100 mL of 1 M HCl was added and the mixture was extracted with hexanes or ethyl acetate (3 × 50 mL). The organic layers were combined, washed with brine and dried over magnesium sulfate. The solids were removed by filtration and solvents were removed *in vacuo*. The derivatives were further purified as described below. The products were then reacted with *N*-bromosuccinimide as reported to form the monomers⁴.

2,5-Di(dec-9-en-1-yl)-3,6-di(thiophen-2-yl)pyrrolo[3,4-*c*]pyrrole-1,4(2*H*,5*H*)-dione (C10H19-DPP): The obtained crude product was purified by column chromatography with hexanes: dichloromethane (1:1 to 1:3) as the eluent ($R_f \sim 0.58$ with hexanes : dichloromethane 1:1). Purple solid was obtained from precipitation into methanol as the product (1.18 g, 40.1%). ¹H NMR (400 MHz, CDCl₃) δ 8.92 (d, $J = 3.9$ Hz, 2H), 7.63 (d, $J = 5.0$ Hz, 2H), 7.28 (t, $J = 4.4$ Hz, 2H), 5.80 (m, 2H), 4.95 (m, 4H), 4.06 (t, $J = 7.9$ Hz, 4H), 2.02 (m, 4H), 1.74 (m, 4H), 1.43 – 1.24 (m, 20H). ¹³C NMR (100 MHz, CDCl₃) δ 161.27, 139.92, 139.07, 135.15, 130.58, 129.66, 128.50, 114.05, 107.58, 42.10, 33.66, 29.83, 29.23, 29.07, 28.90, 28.76, 26.73. MS (ESI, M+H⁺) for C₃₄H₄₅N₂O₂S₂: *calc'd* 577.2917 *found* 577.2910.

2,5-Bis(2-(2-(2-ethoxyethoxy)ethoxy)ethyl)-3,6-di(thiophen-2-yl)pyrrolo[3,4-*c*]pyrrole-1,4(2*H*,5*H*)-dione (TEG-DPP): The crude product was purified by column chromatography with dichloromethane: acetone (10:1 to 5:1) as the eluent ($R_f \sim 0.19$ with dichloromethane : acetone 10:1). Purple needle crystals were obtained from recrystallization from dichloromethane and hexanes as the final product. (1.56 g, 49.1%) ¹H NMR (300 MHz, CDCl₃) δ 8.71 (d, $J = 3.9$ Hz, 2H), 7.58 (d, $J = 5.0$ Hz, 2H), 7.19 (dd, $J_1 = 5.1$ Hz, $J_2 = 3.9$ Hz, 2H), 4.20 (t, $J = 6.2$ Hz, 4H), 3.73 (t, $J = 6.2$ Hz, 4H), 3.68 – 3.43 (m, 20H), 1.12 (t, $J = 7.0$ Hz, 6H). ¹³C NMR (100 MHz, CDCl₃) δ 161.30, 140.20, 134.65, 130.72, 129.53, 128.27, 107.69, 70.59, 70.54, 70.37, 69.66, 68.78, 66.44, 41.71, 15.03. MS (ESI, M+Na⁺) for C₃₀H₄₀N₂O₈S₂Na: *calc'd* 643.2118 *found* 643.2138.

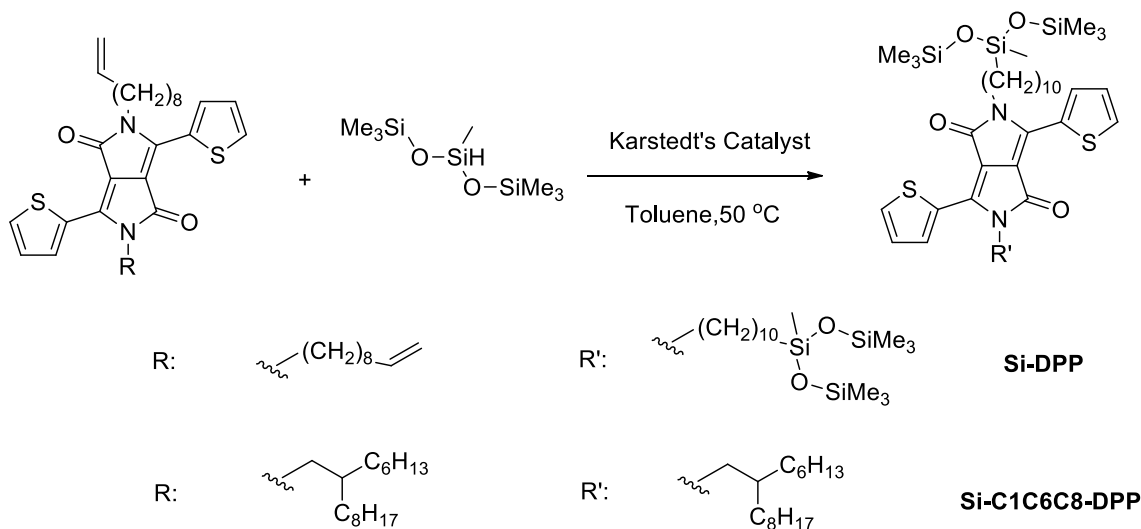
2-(2-(2-(2-Ethoxyethoxy)ethoxy)ethyl)-5-(2-hexyldecyl)-3,6-di(thiophen-2-yl)pyrrolo[3,4-*c*]pyrrole-1,4(2*H*,5*H*)-dione (C1C6C8-TEG-DPP): The crude product was purified by column chromatography with dichloromethane: acetone (10:1) as the eluent ($R_f \sim 0.38$). (24.6%) ¹H NMR (400 MHz, CDCl₃) δ 8.81 (d, $J = 4.0$ Hz, 1H), 8.78 (d, $J = 3.9$ Hz, 1H), 7.61 (m, 2H), 7.24 (m, 2H), 4.27 (t, $J = 6.4$ Hz, 2H), 3.99 (d, $J = 7.7$ Hz, 2H), 3.78 (t, $J = 6.3$ Hz, 2H), 3.68 – 3.42 (m, 10H), 1.87 (m, 1H), 1.31 – 1.12 (m, 27H), 0.83 (m, 6H). ¹³C NMR (100 MHz, CDCl₃) δ 161.63, 161.35, 140.55, 139.99, 135.03, 134.71, 130.56, 130.51, 129.64, 129.63, 128.31, 128.27, 107.94, 107.62, 70.61, 70.43, 69.70, 68.85, 66.49, 46.04, 41.72, 37.61, 31.76, 31.65, 31.02, 29.88, 29.55, 29.38, 29.16, 26.04, 22.52, 15.04, 13.99. MS (ESI, M+H⁺) for C₃₈H₅₇N₂O₅S₂: *calc'd* 685.3709 *found* 685.3705.

2-(Dec-9-en-1-yl)-5-(2-hexyldecyl)-3,6-di(thiophen-2-yl)pyrrolo[3,4-*c*]pyrrole-1,4(2*H*,5*H*)-dione (C10H19-C1C6C8-DPP): The product was purified with the same method of C10H19-DPP ($R_f \sim 0.72$ for hexanes : dichloromethane 1:1). ¹H NMR (400 MHz, CDCl₃) δ 8.95 (dd, $J = 4.0, 0.8$ MHz, 1H), 8.85 (dd, $J = 4.0, 0.8$ MHz, 1H), 7.67 – 7.59 (m, 2H), 7.32 – 7.23 (m, 2H), 5.80 (m, 1H), 5.03 – 4.88 (m, 2H), 4.07 (t, $J = 7.8$ Hz, 2H), 4.01 (d, $J = 7.7$ Hz, 2H), 2.03 (m, 2H), 1.89 (m, 1H), 1.75 (m, 2H), 1.48 – 1.10 (m, 34H), 0.84 (m, 6H). ¹³C NMR (100 MHz, CDCl₃) δ 161.64, 161.27, 140.27, 139.96, 139.06, 135.28, 134.97, 130.50, 130.41, 129.74, 129.68, 128.53, 128.26,

114.05, 107.96, 107.47, 46.09, 42.12, 37.63, 33.67, 31.77, 31.66, 31.07, 29.87, 29.56, 29.38, 29.18, 29.07, 28.77, 26.75, 26.08, 22.54, 14.00. MS (ESI, $M+H^+$) for $C_{40}H_{59}N_2O_2S_2$: *calc'd* 663.4012 *found* 663.4019.

3,6-Bis(5-bromothiophen-2-yl)-2,5-bis(2-(2-(2-ethoxyethoxy)ethoxy)ethyl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione (TEG-DPP-Br₂): The product was purified by column chromatography with dichloromethane: acetone (20:1 to 10:1) as the eluent ($R_f \sim 0.40$ for dichloromethane : acetone 10:1, 37%). ¹H NMR (400 MHz, $CDCl_3$) δ 8.49 (d, $J = 4.2$ Hz, 2H), 7.21 (d, $J = 4.1$ Hz, 2H), 4.17 (t, $J = 4.5$ Hz, 4H), 3.77 (t, $J = 4.5$ Hz, 4H), 3.64 – 3.47 (m, 20H), 1.19 (t, $J = 5.1$ Hz, 6H). ¹³C NMR (100 MHz, $CDCl_3$) δ 160.99, 139.26, 134.75, 131.25, 130.99, 119.23, 107.81, 70.76, 70.70, 70.50, 69.80, 68.92, 66.62, 42.28, 15.28. MS (ESI, $M+H^+$) for $C_{30}H_{39}Br_2N_2O_8S_2$: *calc'd* 777.0509 *found* 777.0520.

3,6-Bis(5-bromothiophen-2-yl)-2-(2-(2-(2-ethoxyethoxy)ethoxy)ethyl)-5-(2-hexyldecyl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione (C1C6C8-TEG-DPP-Br₂): The product was purified by the same method of TEG-DPP-Br₂ ($R_f \sim 0.78$ in dichloromethane : acetone 10:1, 24.6%). ¹H NMR (400 MHz, $CHCl_3$) δ 8.57 (d, $J = 4.2$ Hz, 1H), 8.52 (d, $J = 4.2$ Hz, 1H), 7.21 (d, $J = 4.4$ Hz, 1H), 7.19 (d, $J = 4.4$ Hz, 1H), 4.17 (t, $J = 6.0$ Hz, 2H), 3.90 (d, $J = 7.7$ Hz, 2H), 3.77 (t, $J = 6.0$ Hz, 2H), 3.65 – 3.43 (m, 12H), 1.85 (m, 1H), 1.27 – 1.16 (m, 25H), 0.85 (m, 6H). ¹³C NMR (100 MHz, $CDCl_3$) δ 161.30, 161.07, 139.48, 139.12, 135.12, 134.79, 131.29, 131.05, 131.00, 119.04, 108.01, 107.72, 70.69, 70.63, 70.44, 69.72, 68.86, 66.52, 46.17, 42.12, 37.66, 31.77, 31.66, 31.01, 29.87, 29.55, 29.40, 29.17, 26.03, 22.56, 15.05, 14.01. MS (ESI, $M+H^+$) for $C_{38}H_{55}Br_2N_2O_5S_2$: *calc'd* 841.1914 *found* 841.1928.

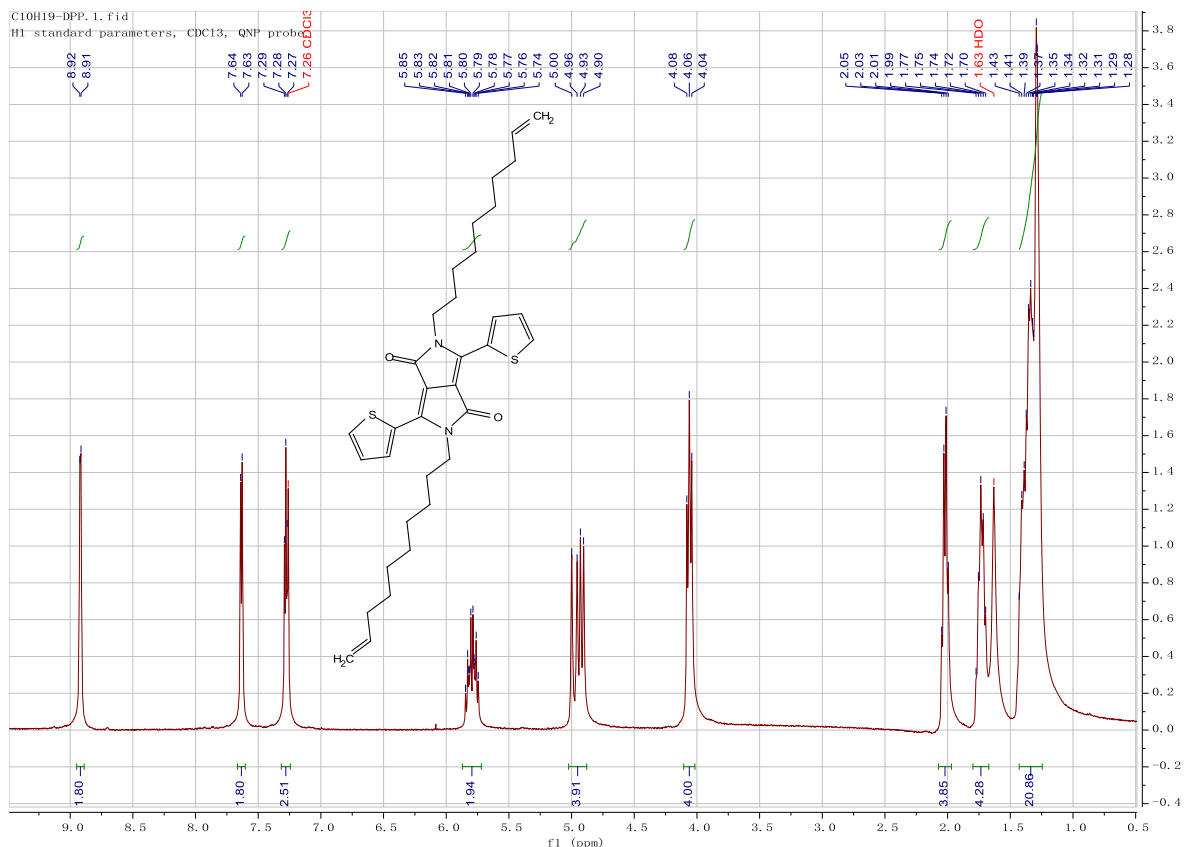
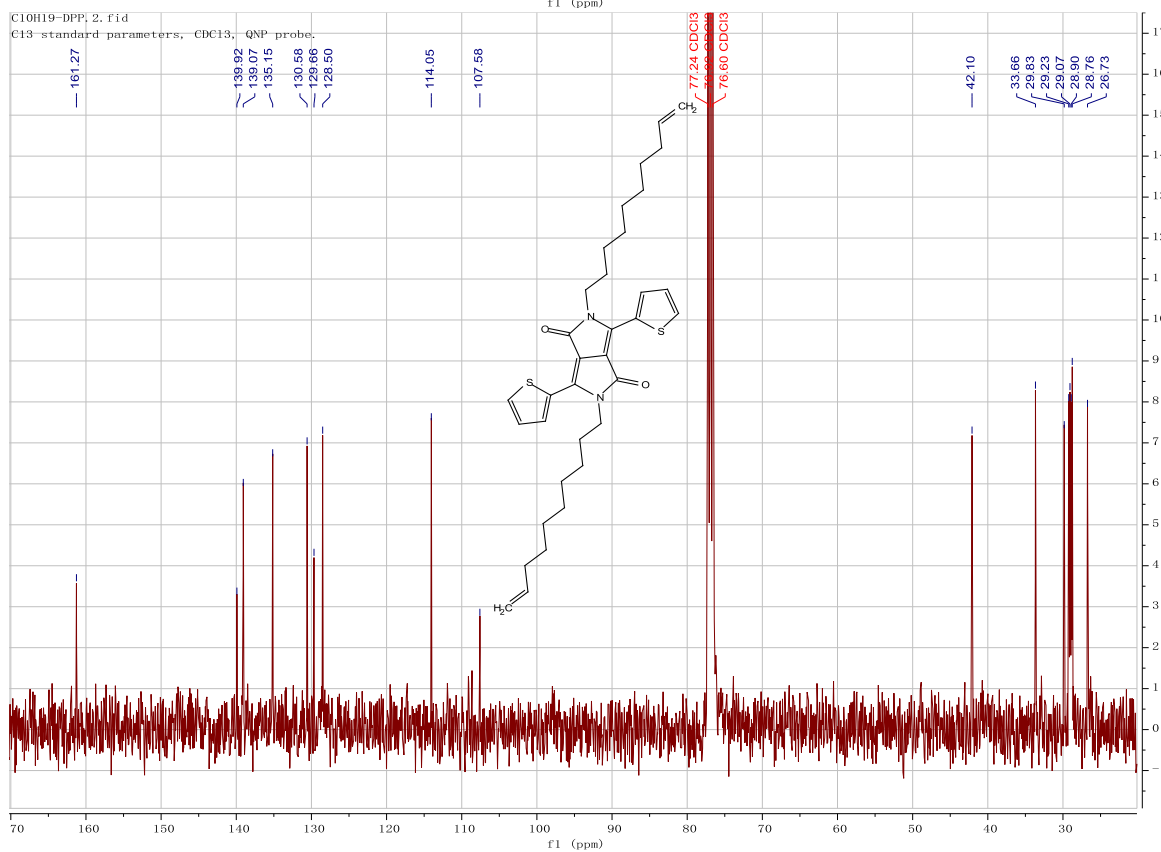


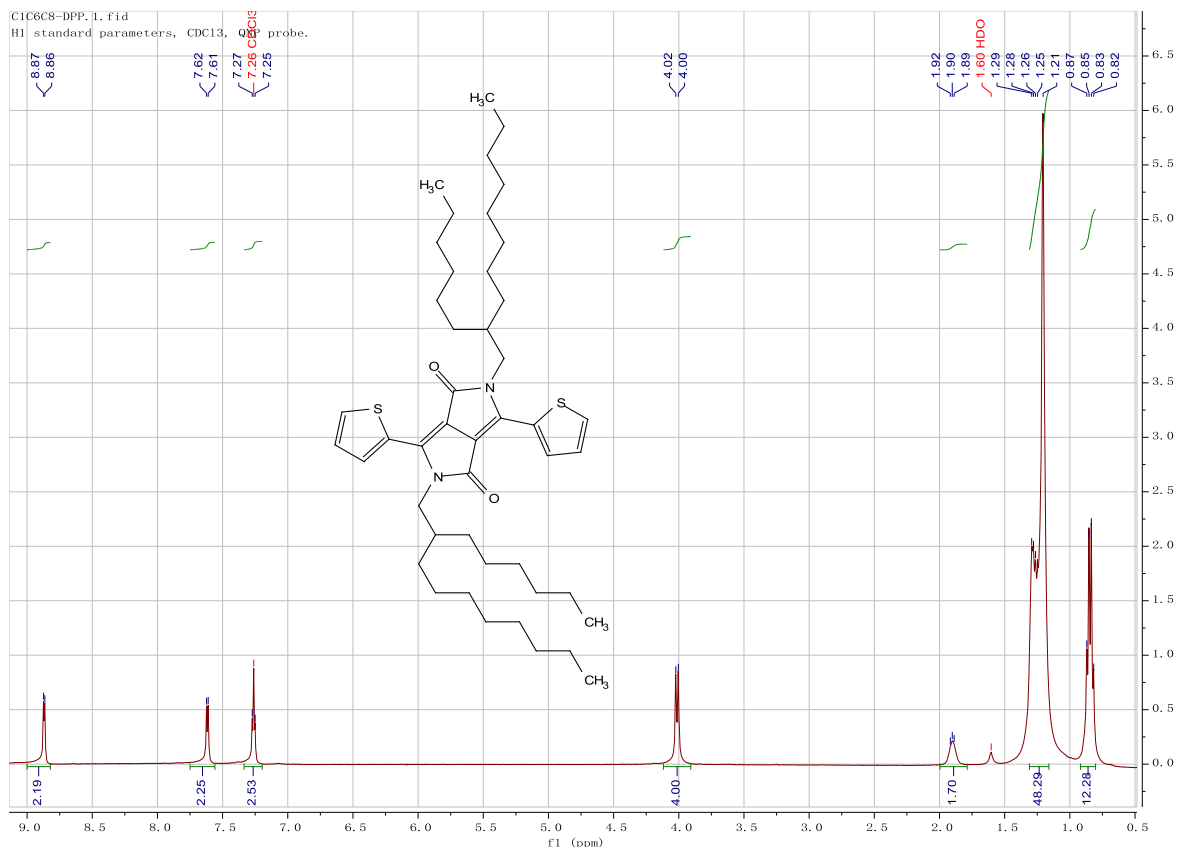
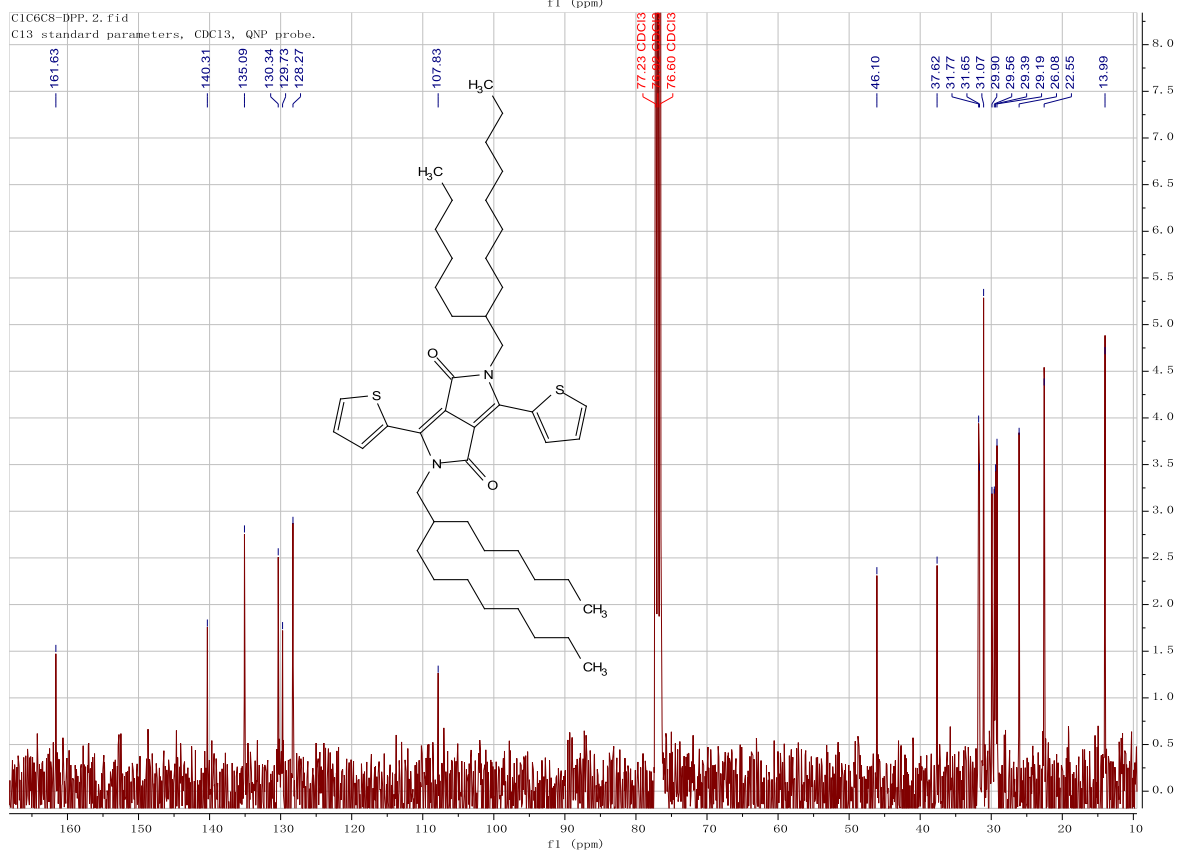
2,5-Bis(10-(1,1,1,3,5,5,5-heptamethyltrisiloxan-3-yl)decyl)-3,6-di(thiophen-2-yl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione (Si-DPP): The DPP derivatives with siloxane-terminated side chains were synthesized based on the procedure previously reported (94.0%).^{5, 6} ¹H NMR (400 MHz, $CDCl_3$) δ 8.93 (d, $J = 3.9$ Hz, 2H), 7.62 (d, $J = 5.1$ Hz, 2H), 7.28 – 7.25 (m, 2H), 4.05 (t, $J = 8.0$ Hz, 4H), 1.72 (m, 4H), 1.45 – 1.18 (m, 28H), 0.43 (t, $J = 7.9$ Hz, 4H), 0.03 (s, 36H), -0.01 (s, 6H). ¹³C NMR (100 MHz, $CDCl_3$) δ 161.19, 139.85, 135.17, 130.51, 129.67, 128.47, 107.55, 42.11, 33.13, 29.86, 29.48, 29.42, 29.24, 29.17, 26.80, 22.96, 17.52, 1.77, -0.36. MS (ESI, $M+H^+$) for $C_{48}H_{89}N_2O_6S_2Si_6$: *calc'd* 1021.4772 *found* 1021.4778.

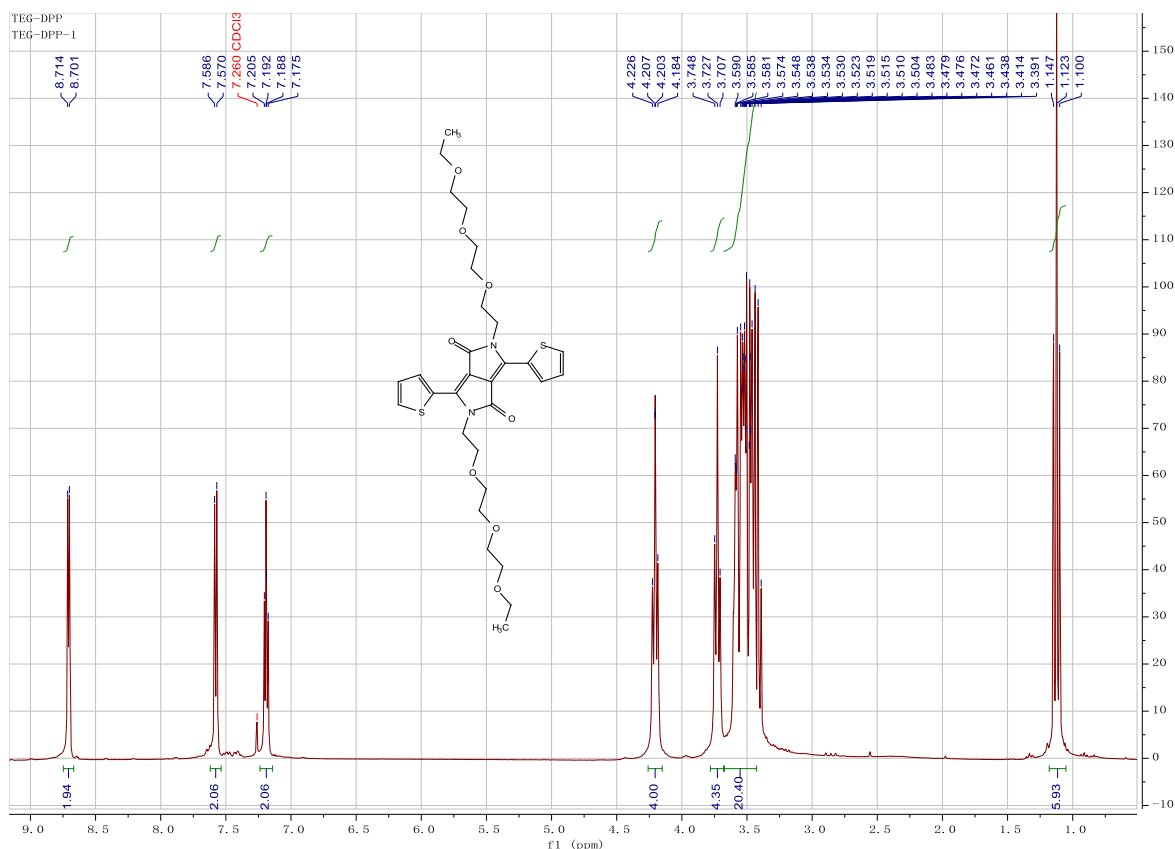
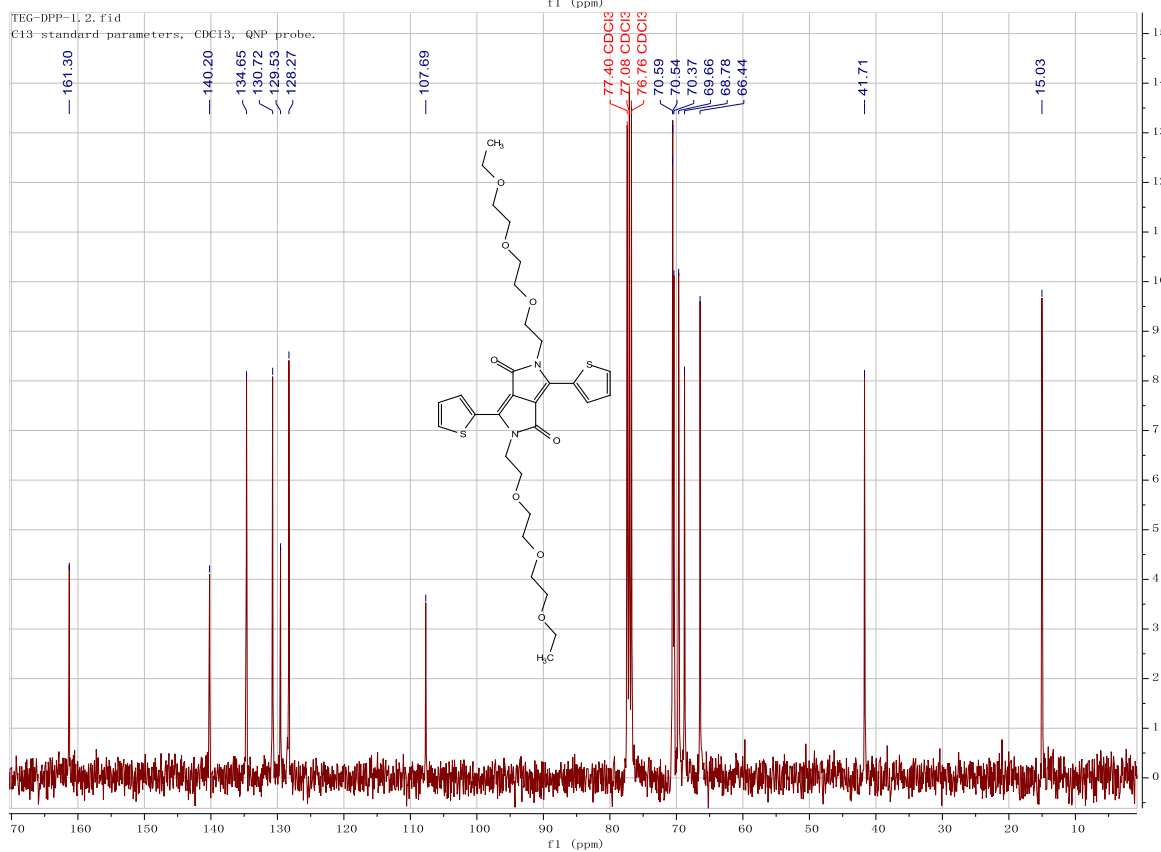
2-(10-(1,1,1,3,5,5,5-Heptamethyltrisiloxan-3-yl)decyl)-5-(2-hexyldecyl)-3,6-di(thiophen-2-yl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione (Si-C1C6C8-DPP): Product was obtained as purple solid (76.1%). ¹H NMR (400 MHz, CDCl₃) δ 8.95 (d, *J* = 3.9 Hz, 1H), 8.86 (d, *J* = 3.9 Hz, 1H), 7.60 (m, 2H), 7.24 (m, 2H), 4.04 (t, *J* = 8.0 Hz, 2H), 3.99 (d, *J* = 7.7 Hz, 2H), 1.88 (m, 1H), 1.74 (m, 2H), 1.40 (m, 2H), 1.30 – 1.16 (m, 36H), 0.83 (m, 6H), 0.44 (t, *J* = 8.0 Hz, 2H), 0.08 (s, 18H), -0.01 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 161.55, 161.17, 140.17, 139.89, 135.30, 135.01, 130.45, 130.35, 129.74, 129.67, 128.48, 128.22, 107.91, 107.42, 46.06, 42.12, 37.61, 33.13, 31.77, 31.66, 31.05, 29.87, 29.57, 29.48, 29.42, 29.38, 29.25, 29.18, 26.81, 26.07, 22.97, 22.55, 22.53, 17.52, 14.01, 1.77, -0.35. MS (ESI, M+H⁺) for C₄₇H₈₁N₂O₄S₂Si₃: *calc'd* 885.4940 *found* 885.4937.

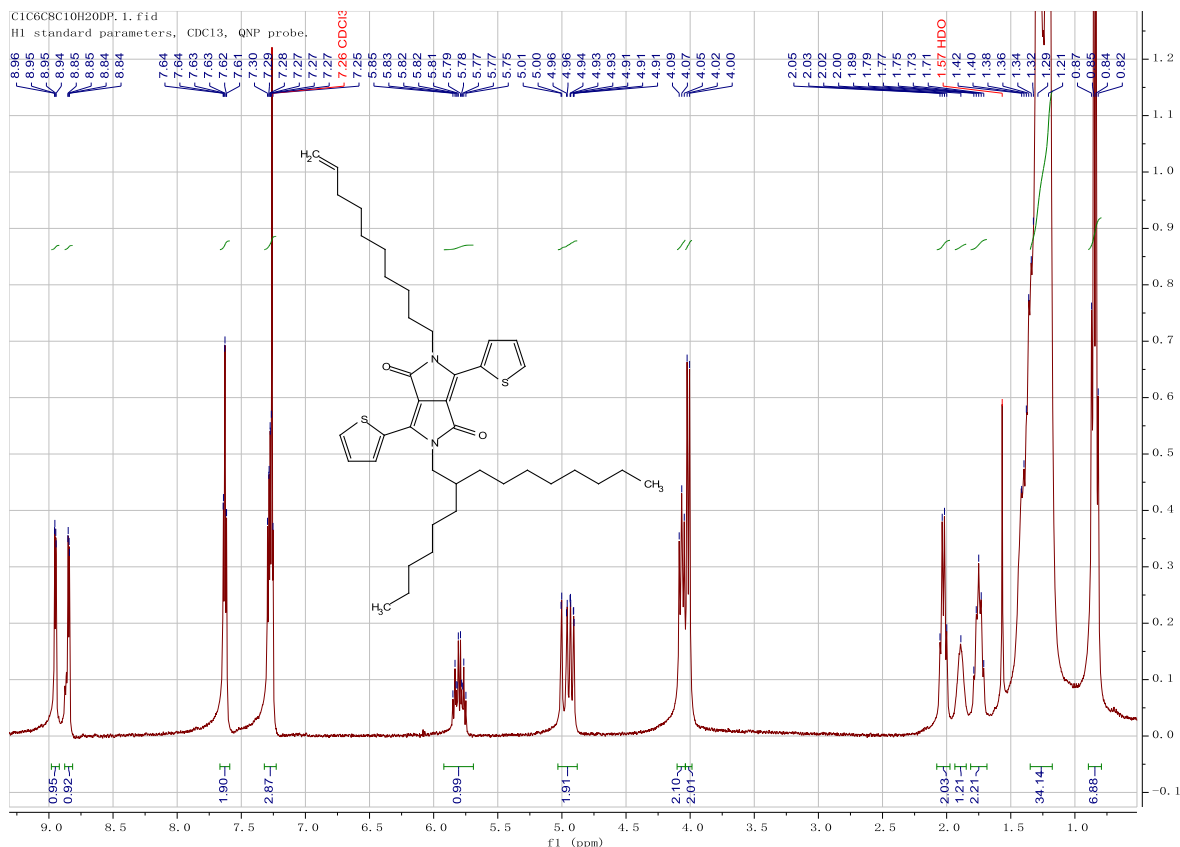
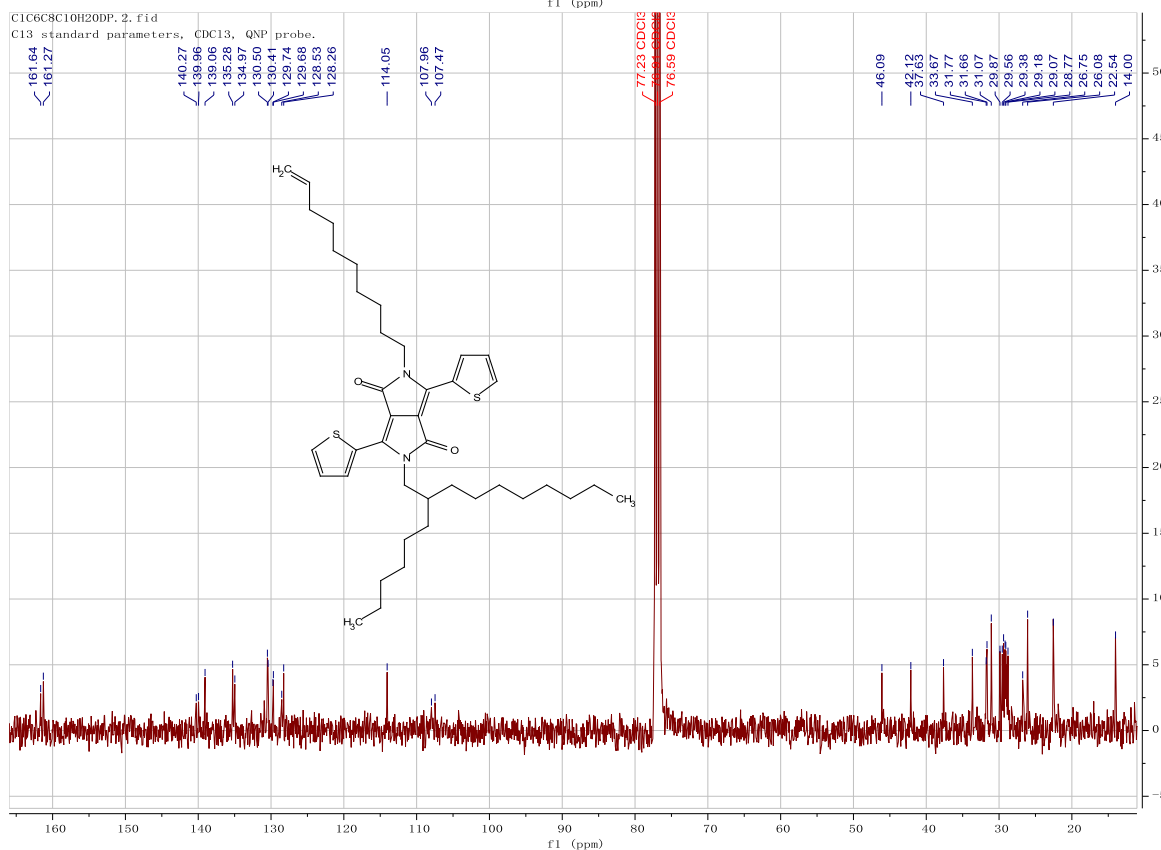
3,6-Bis(5-bromothiophen-2-yl)-2,5-bis(10-(1,1,1,3,5,5,5-heptamethyltrisiloxan-3-yl)decyl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione (Si-DPP-Br₂): (*R_f* ~ 0.80 for hexanes : dichloromethane 1:1, 48%). ¹H NMR (400 MHz, CDCl₃) δ 8.67 (d, *J* = 4.2 Hz, 2H), 7.22 (d, *J* = 4.2 Hz, 2H), 3.97 (t, *J* = 7.9 Hz, 4H), 1.69 (q, *J* = 8.0, 7.6 Hz, 4H), 1.40 – 1.26 (m, 28H), 0.44 (t, *J* = 7.6 Hz, 4H), 0.08 (s, 36H), -0.01 (s, 6H). ¹³C NMR (100 MHz, CDCl₃) δ 160.89, 138.85, 135.25, 131.52, 131.01, 119.02, 107.67, 42.19, 33.14, 29.88, 29.43, 29.25, 29.13, 26.76, 22.97, 17.53, 1.77, -0.37. MS (ESI, M+H⁺) for C₄₈H₈₇Br₂N₂O₆S₂Si₆: *calc'd* 1177.2982 *found* 1177.2977.

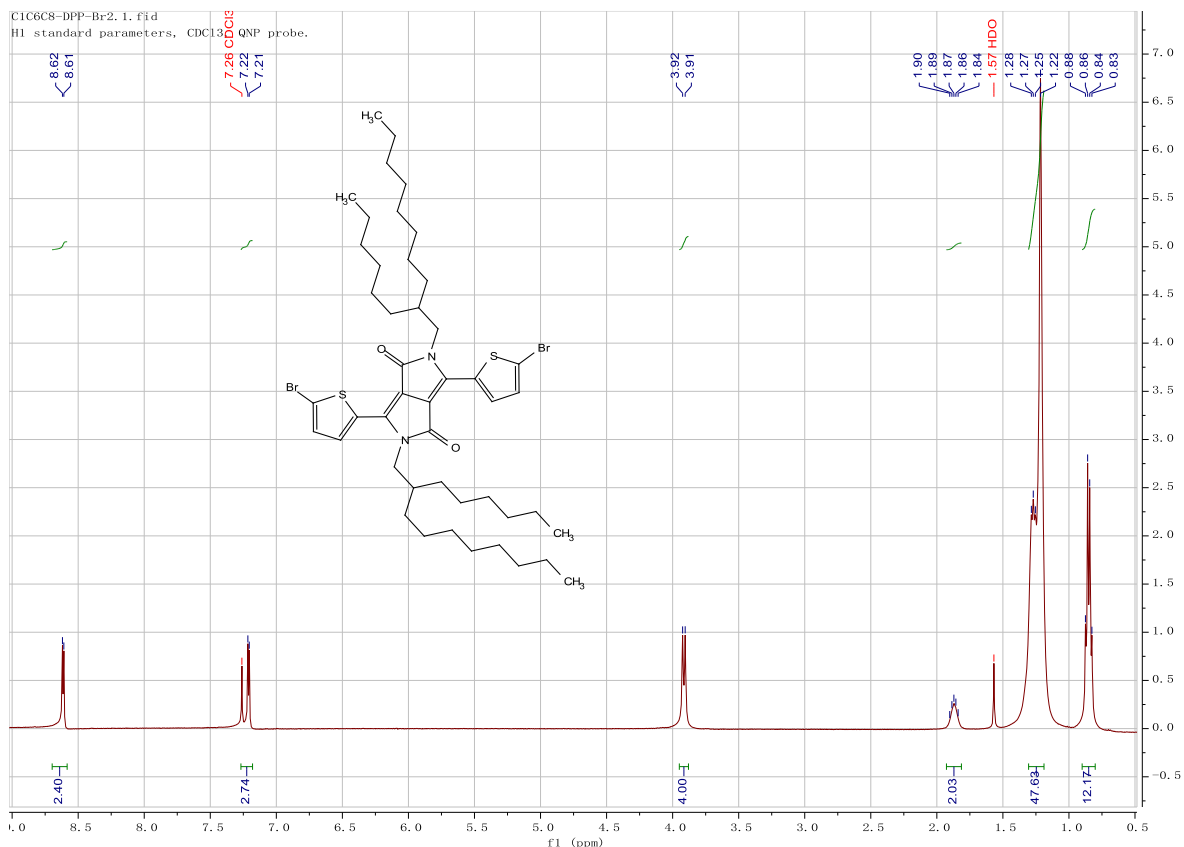
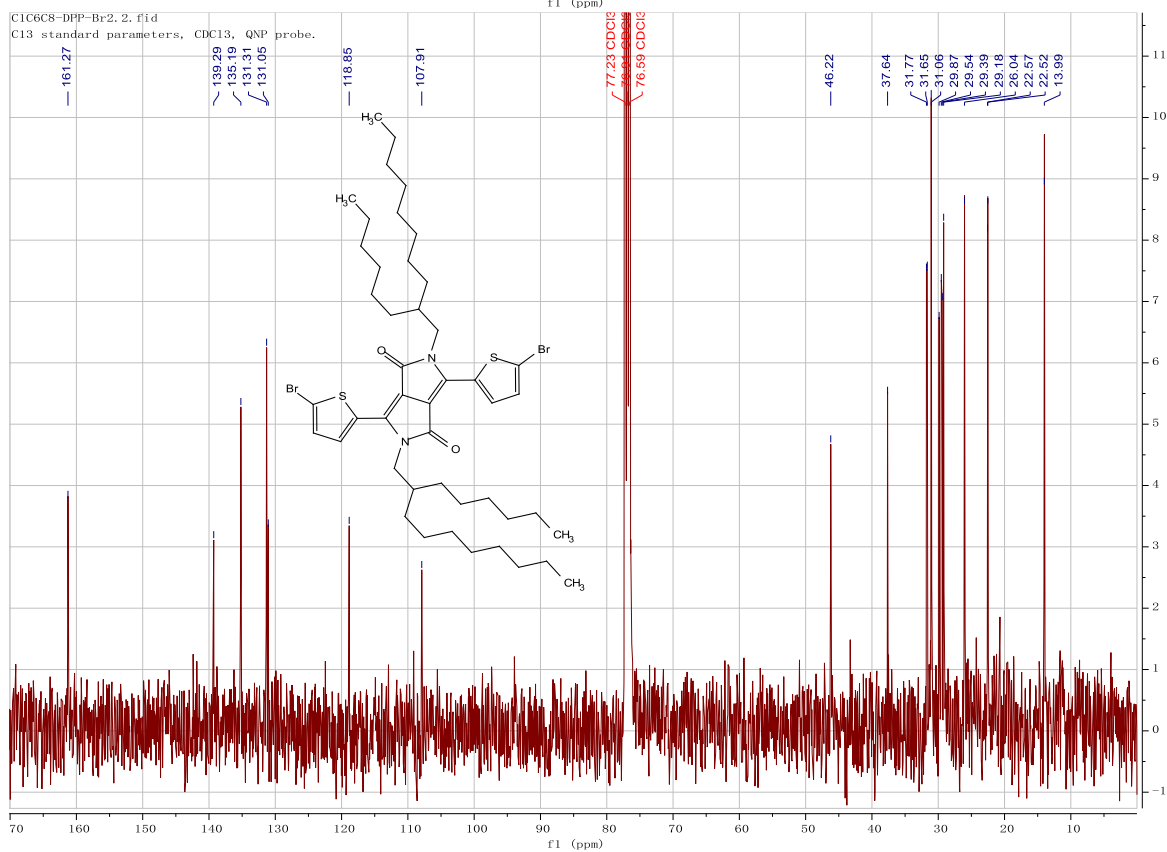
3,6-bis(5-bromothiophen-2-yl)-2-(10-(1,1,1,3,5,5,5-heptamethyltrisiloxan-3-yl)decyl)-5-(2-hexyldecyl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione (Si-C1C6C8-DPP-Br₂): (*R_f* ~ 0.86 in hexanes : dichloromethane 1:1, 34.4%). ¹H NMR (400 MHz, CDCl₃) δ 8.69 (d, *J* = 4.2 Hz, 1H), 8.60 (d, *J* = 4.2 Hz, 1H), 7.22 (m, 2H), 3.97 (t, *J* = 8.0 Hz, 2H), 3.91 (d, *J* = 7.8 Hz, 2H), 1.85 (m, 1H), 1.71 (m, 2H), 1.38 – 1.17 (m, 38H), 0.85 (m, 6H), 0.43 (t, *J* = 7.6 Hz, 2H), 0.08 (s, 18H), -0.01 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 161.26, 160.89, 139.17, 138.95, 135.38, 135.08, 131.55, 131.29, 131.08, 130.99, 119.01, 118.88, 108.02, 107.53, 46.20, 42.20, 37.65, 33.15, 31.78, 31.66, 31.04, 29.86, 29.54, 29.43, 29.39, 29.26, 29.17, 29.13, 26.77, 26.04, 22.98, 22.57, 22.53, 17.53, 14.01, 1.77, -0.36. MS (ESI, M+H⁺) for C₄₇H₇₉Br₂N₂O₄S₂Si₃: *calc'd* 1041.3156 *found* 1041.3170.

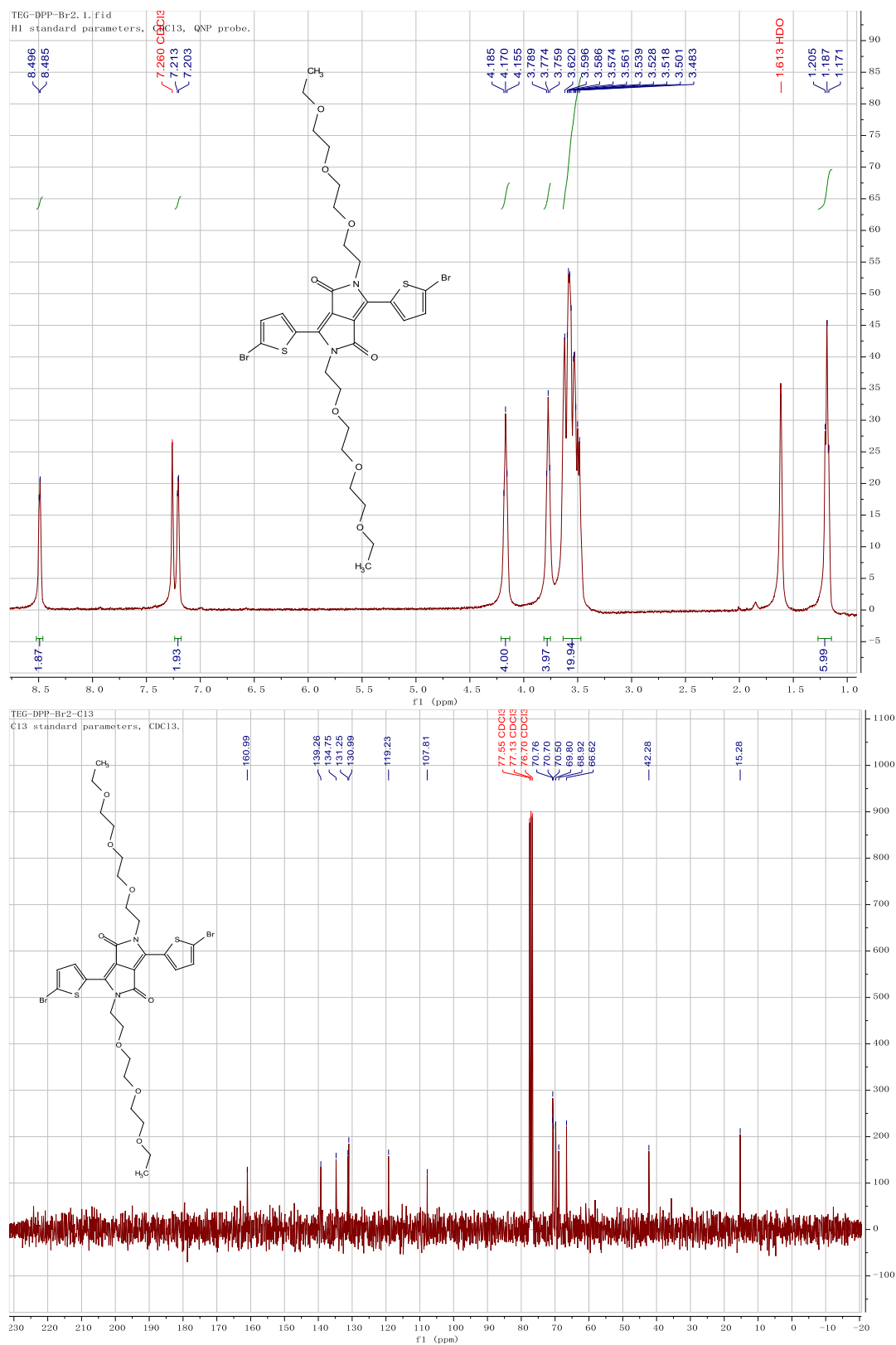


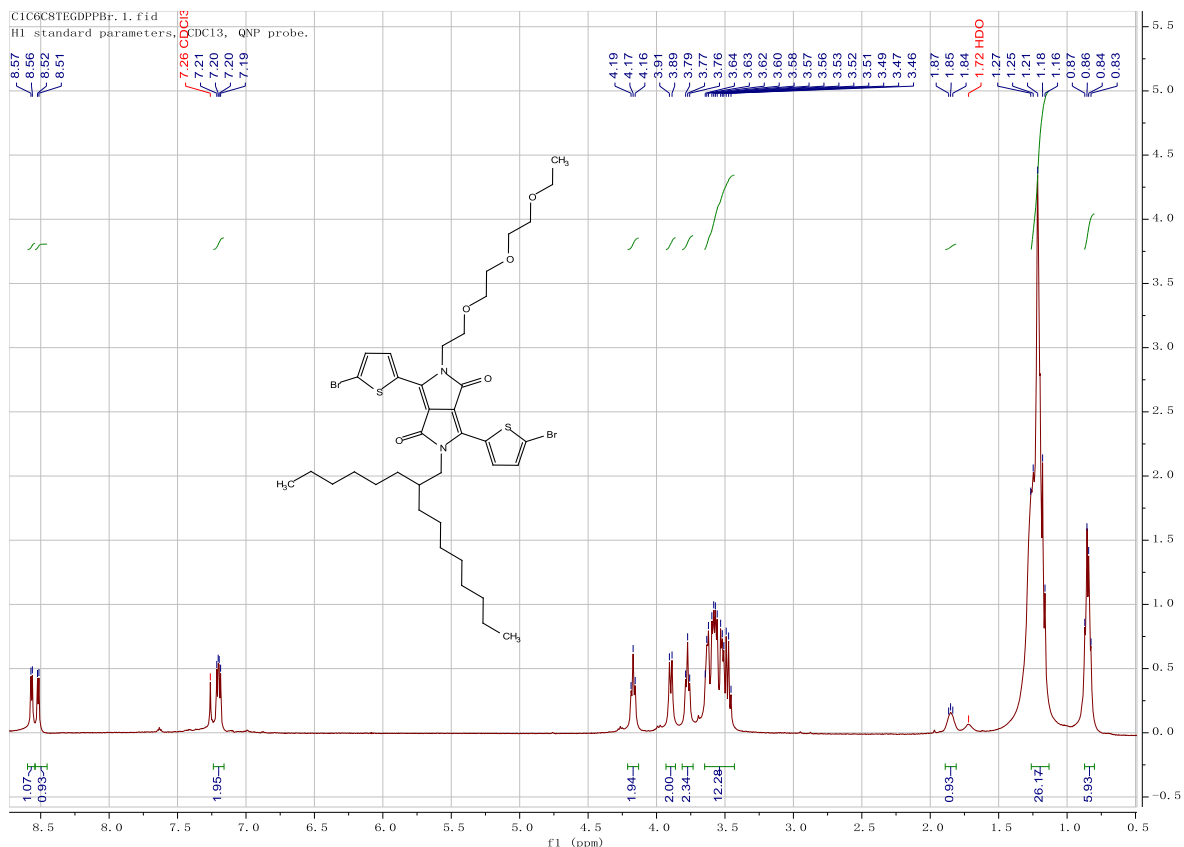
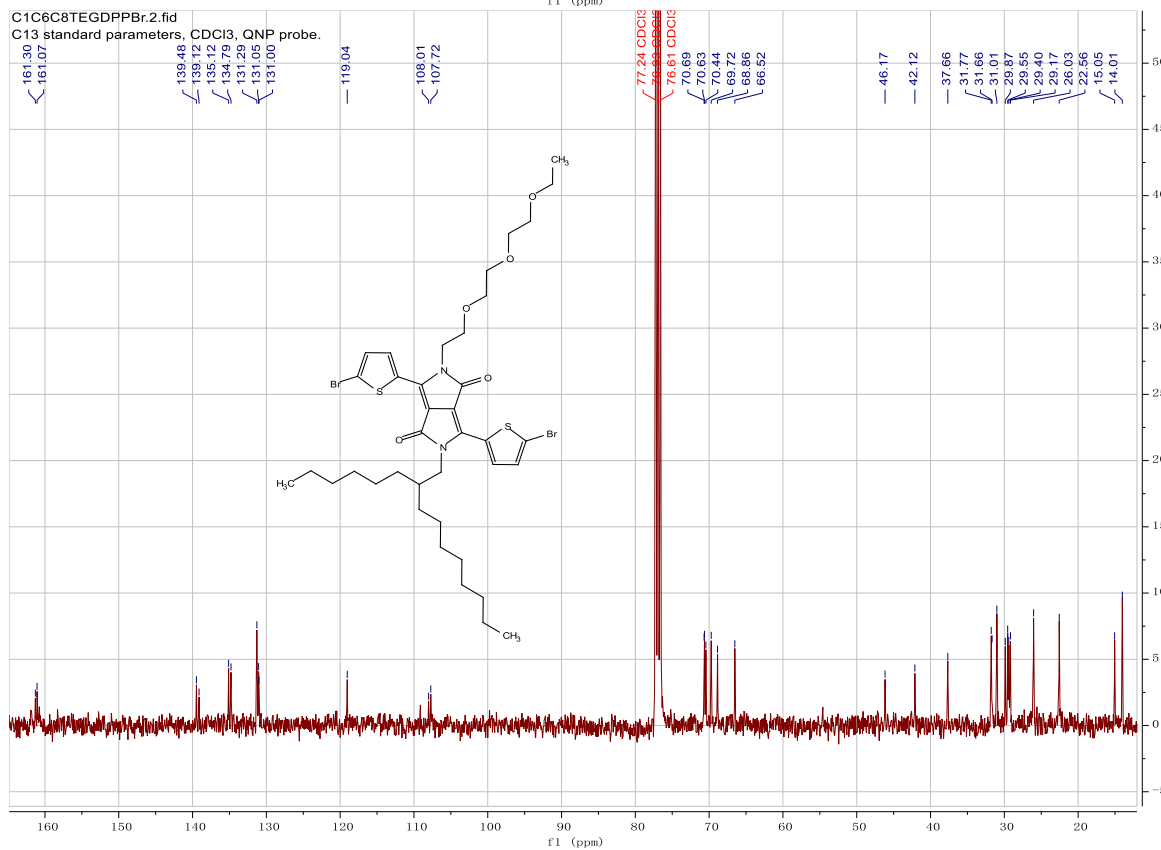


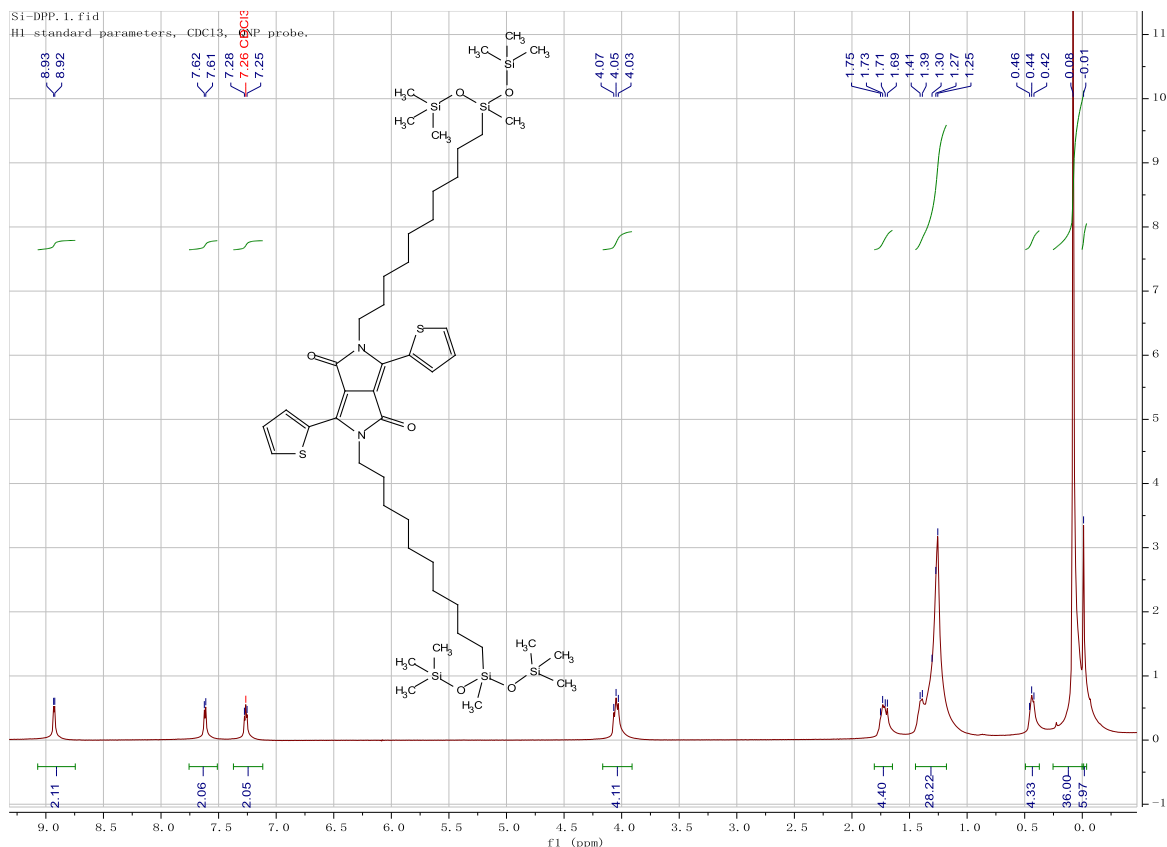
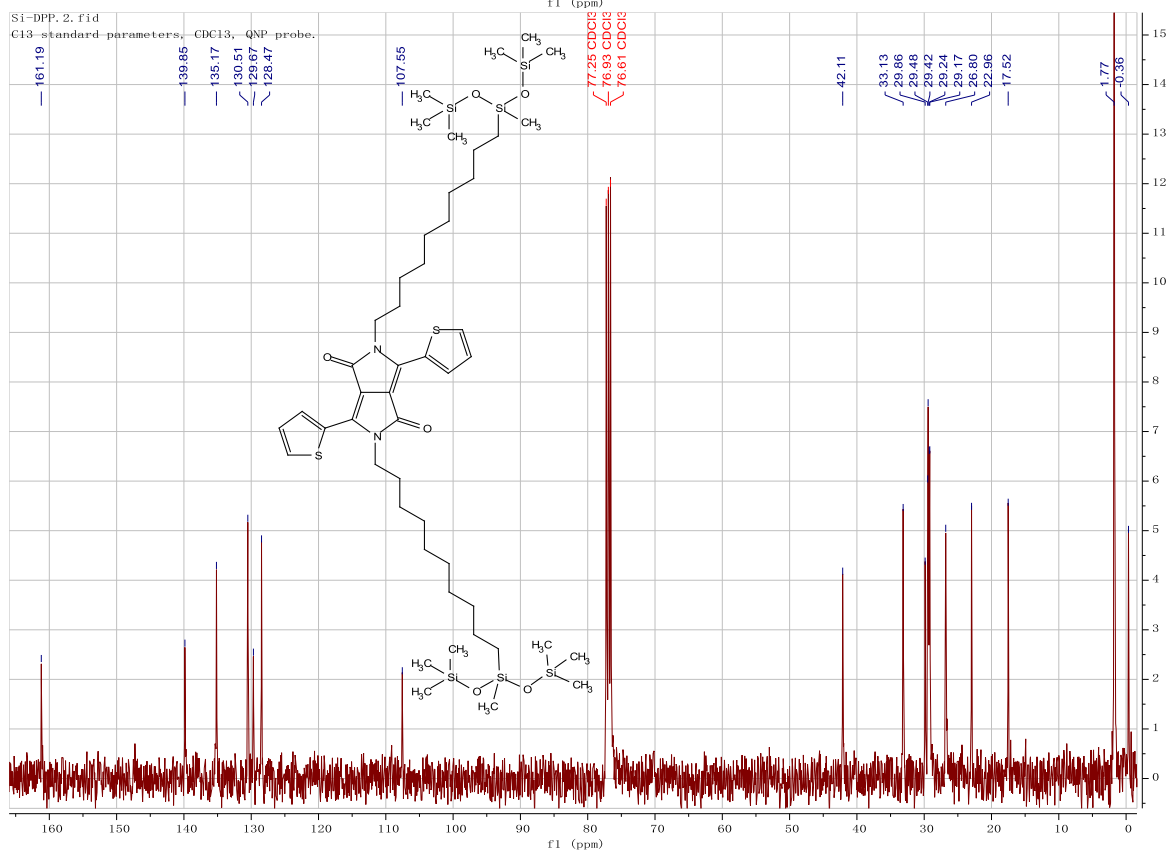


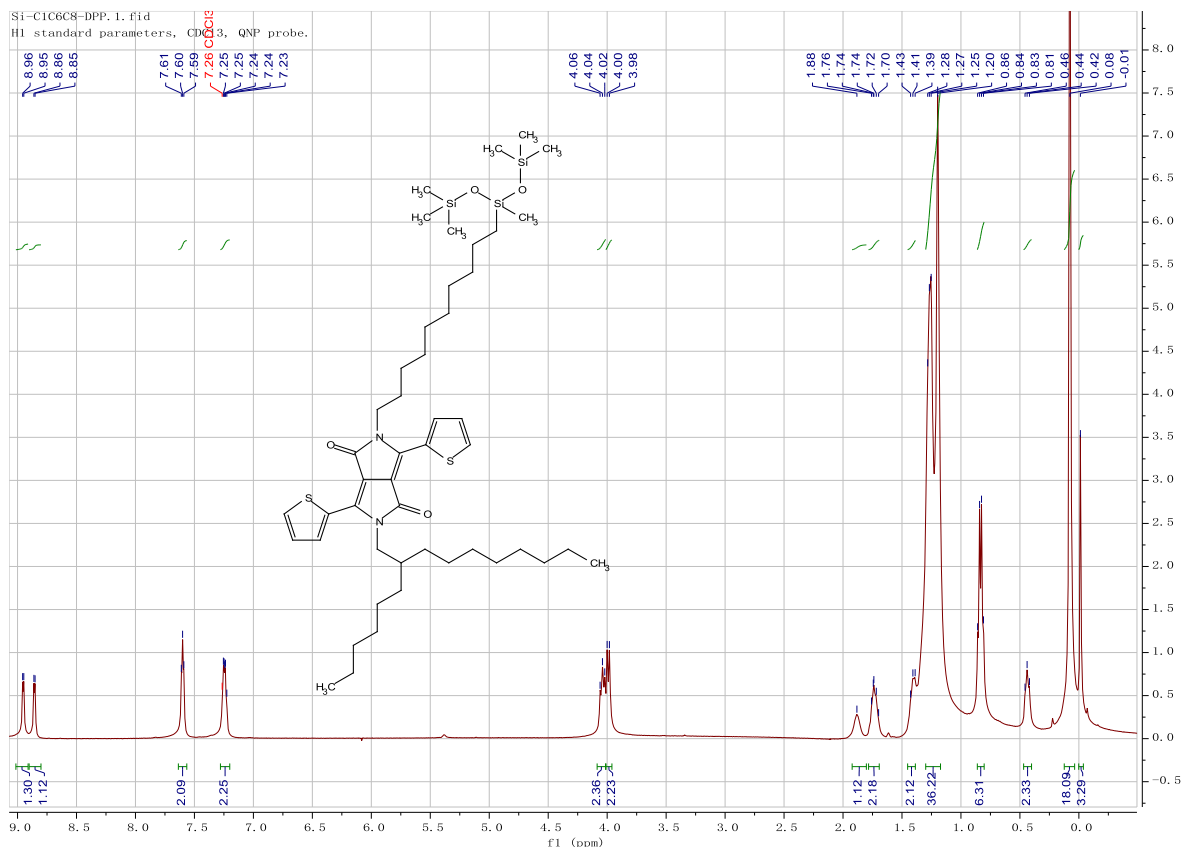
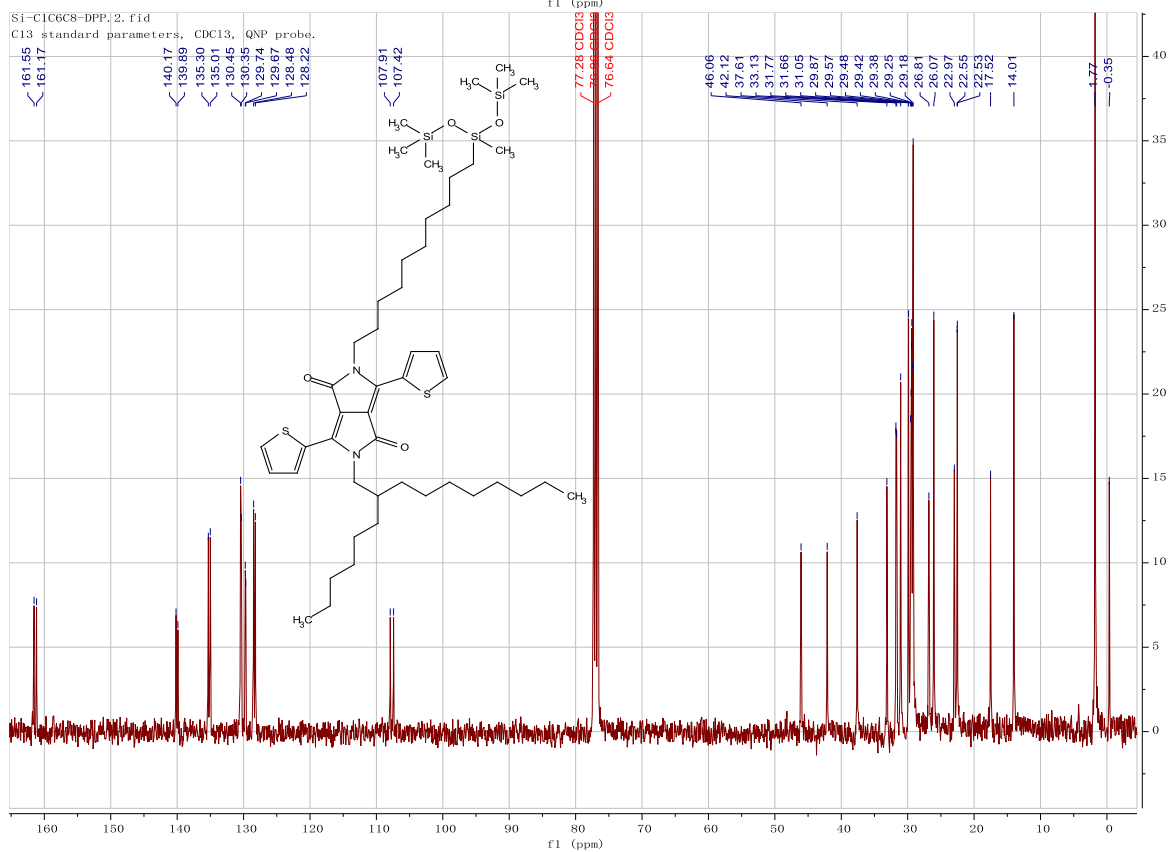


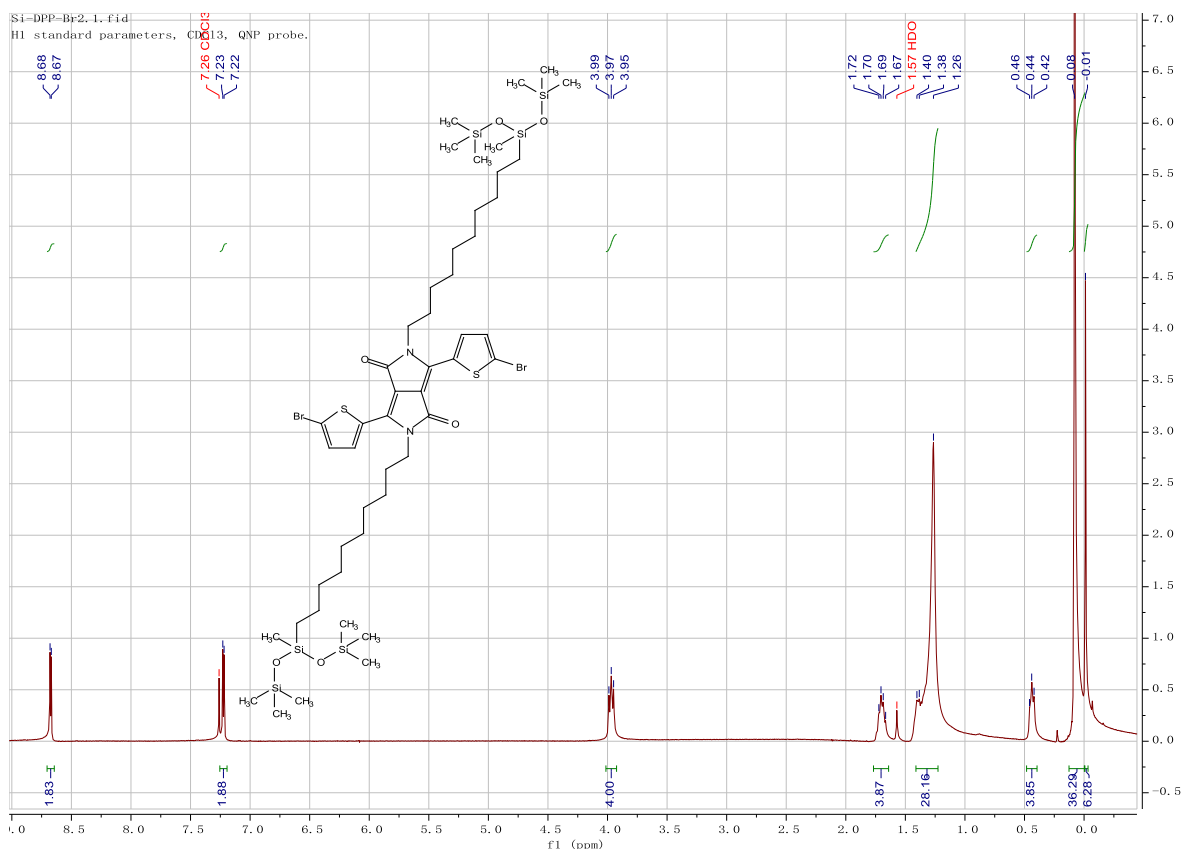
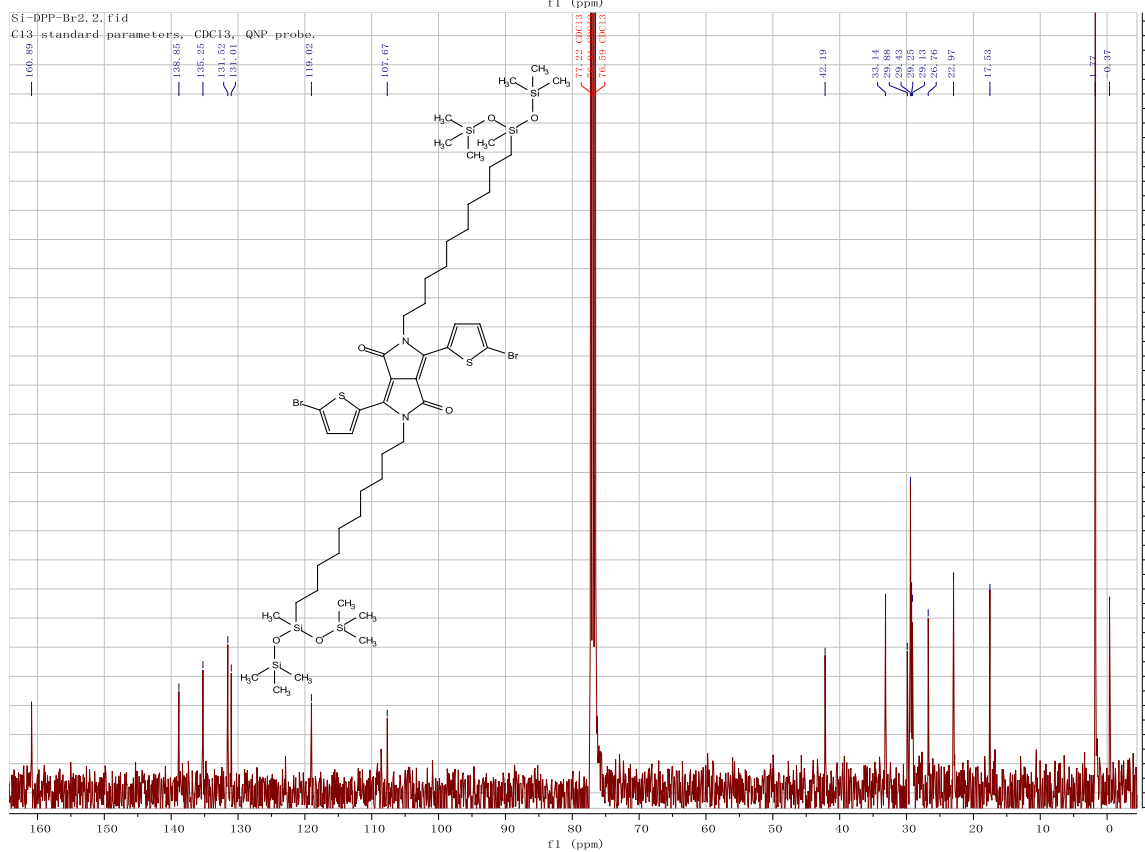


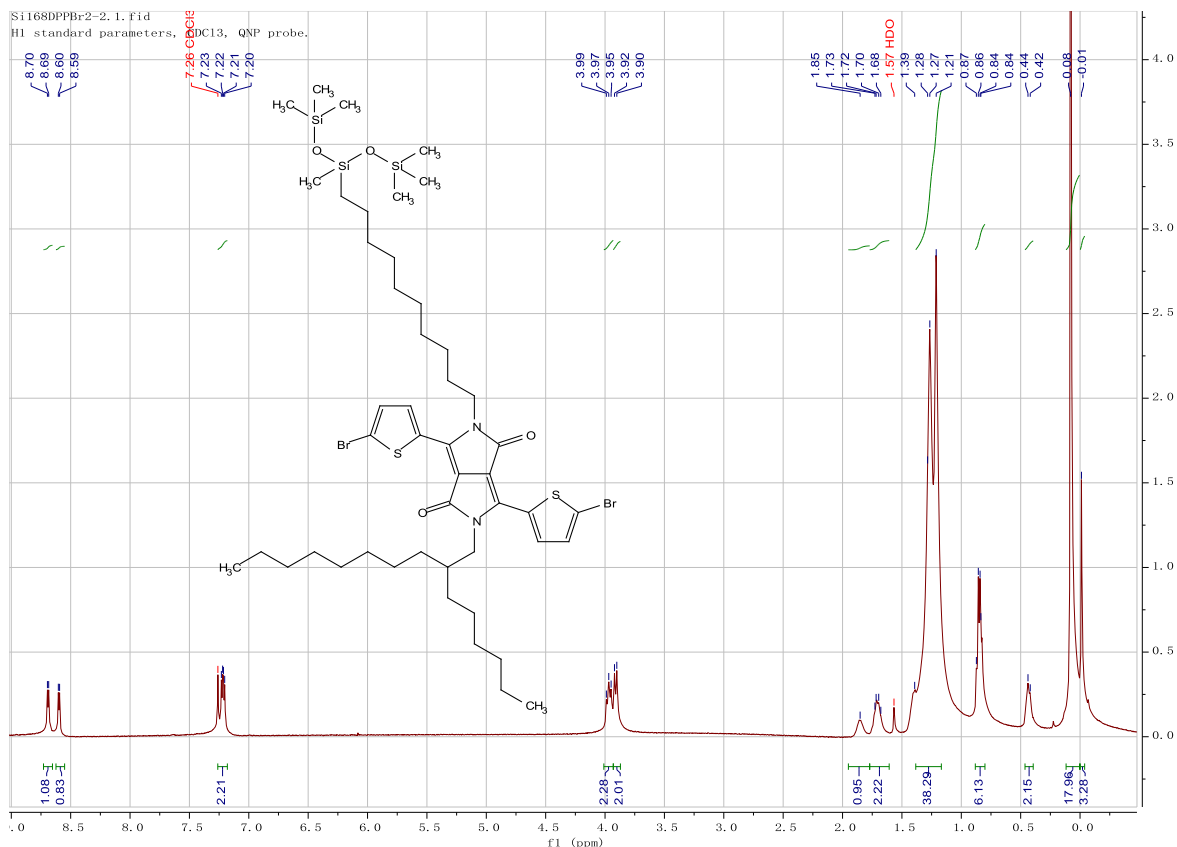
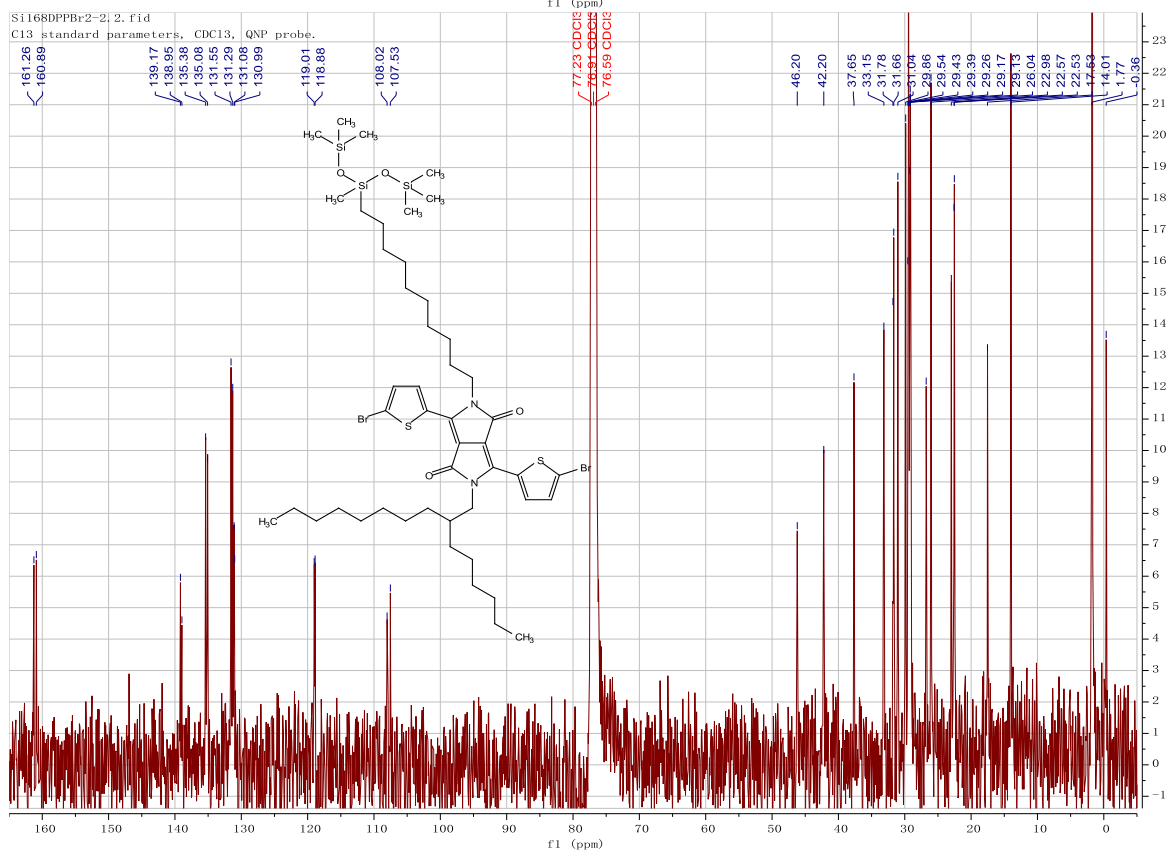


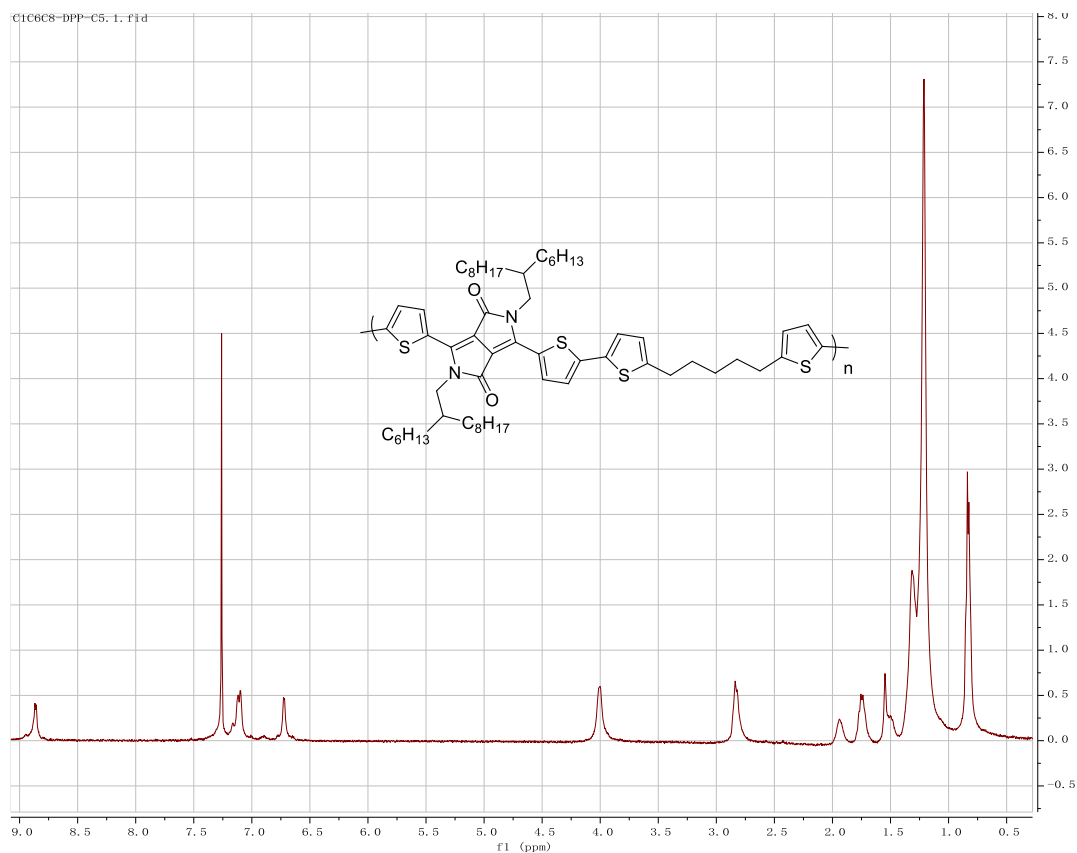
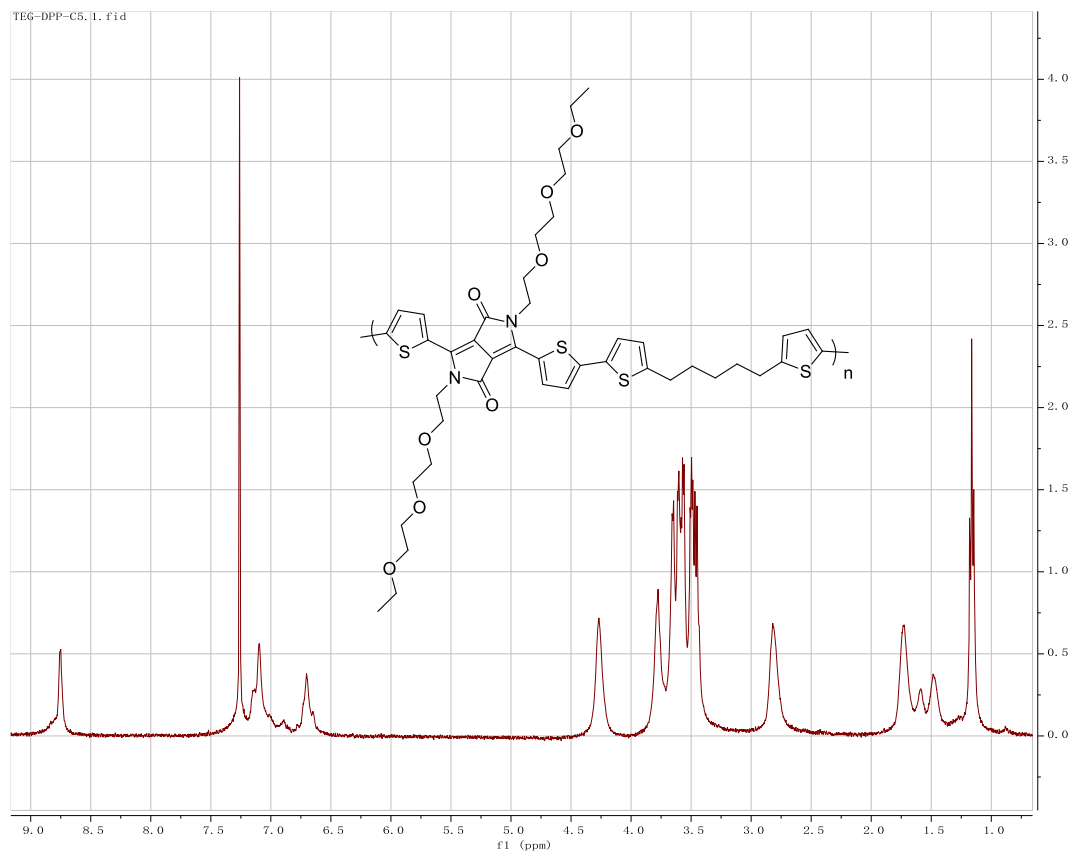


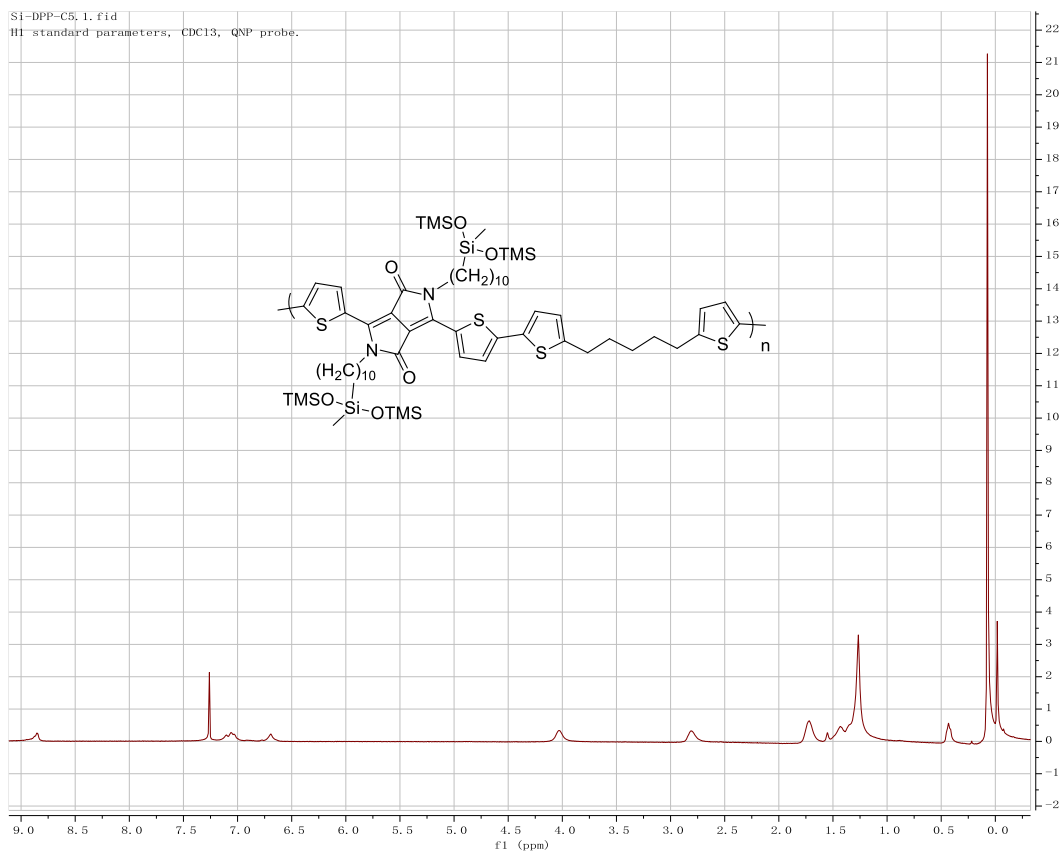




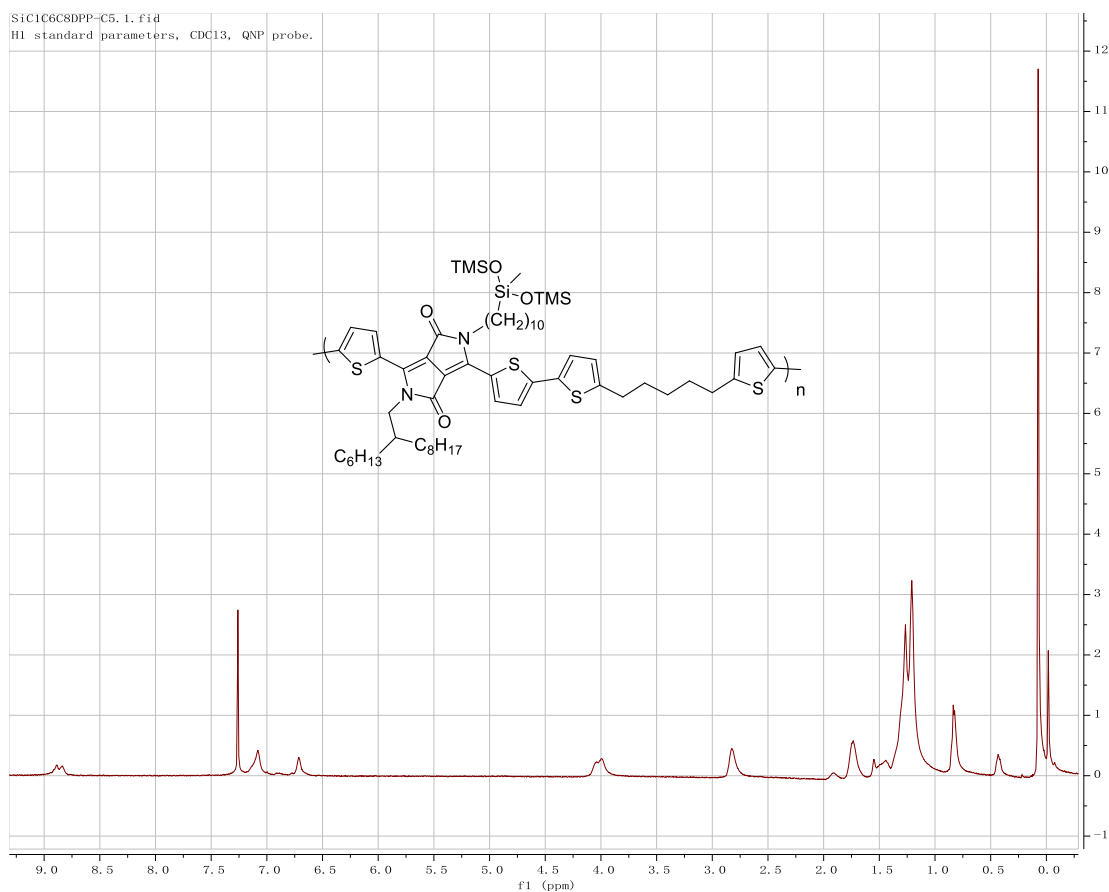








SiC1C6C8DPP-C5.1.fid
 H1 standard parameters, CDCl3, QNP probe.



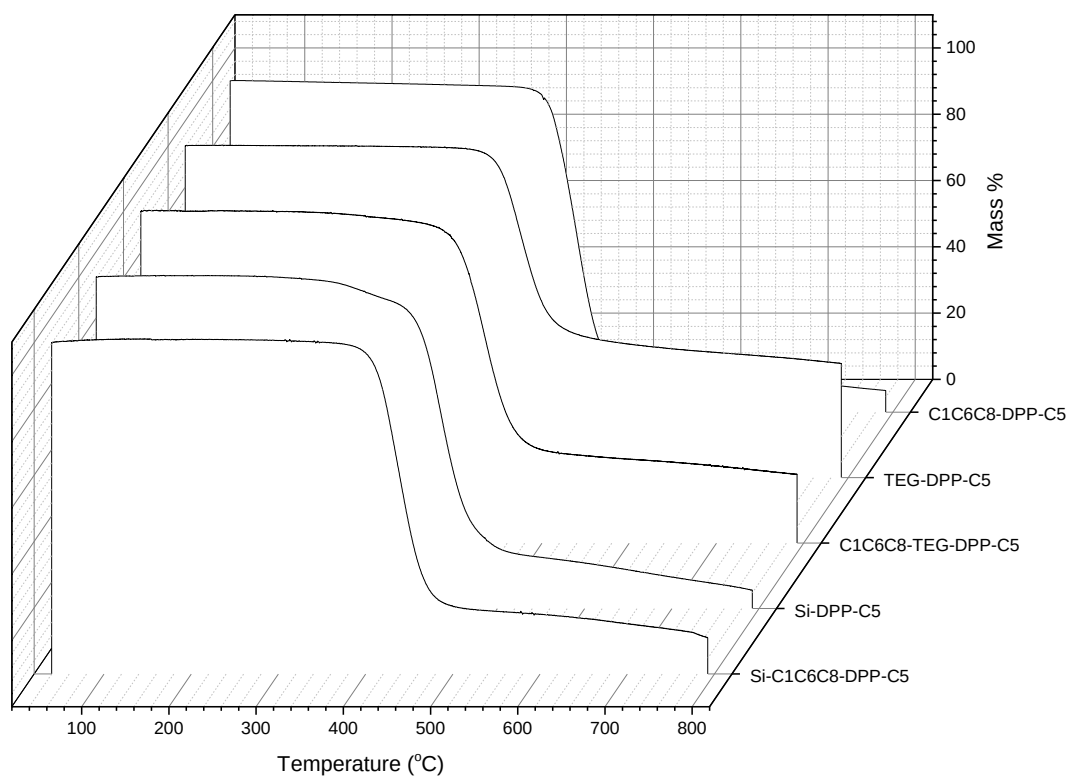
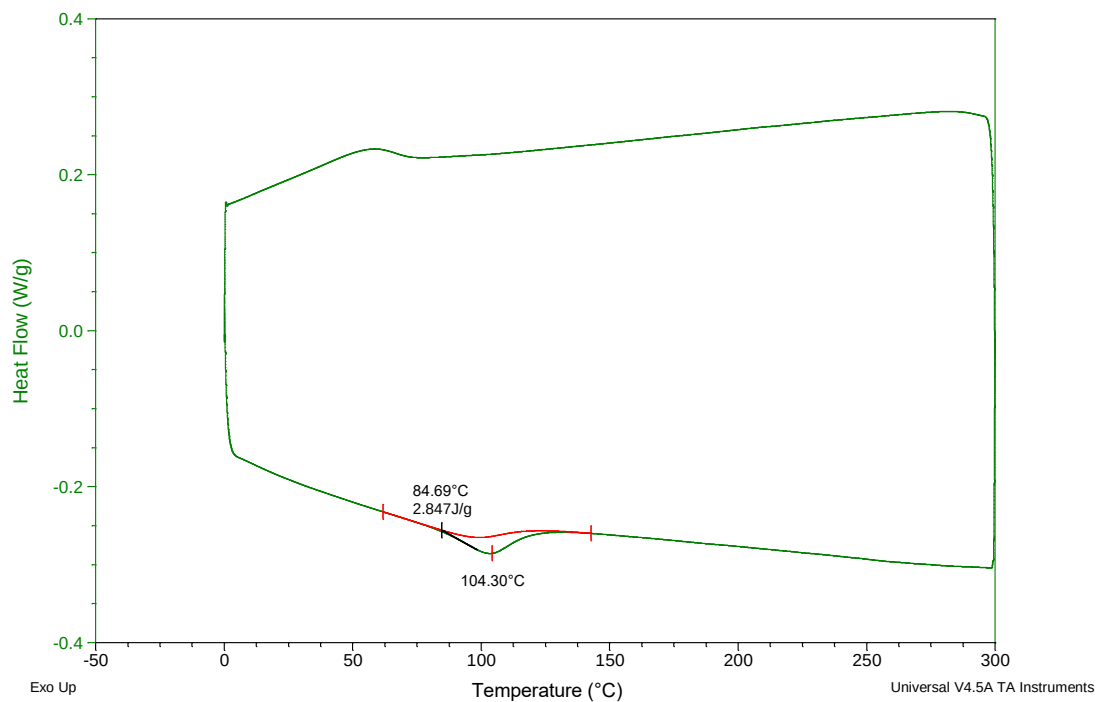


Figure S1. Thermogravimetric analysis (TGA) results of the matrix polymers (10 °C/min under nitrogen atmosphere)

Sample: C1C6C8-TEG-DPP-C5
Size: 4.6000 mg
Method: DPP-Standard

DSC

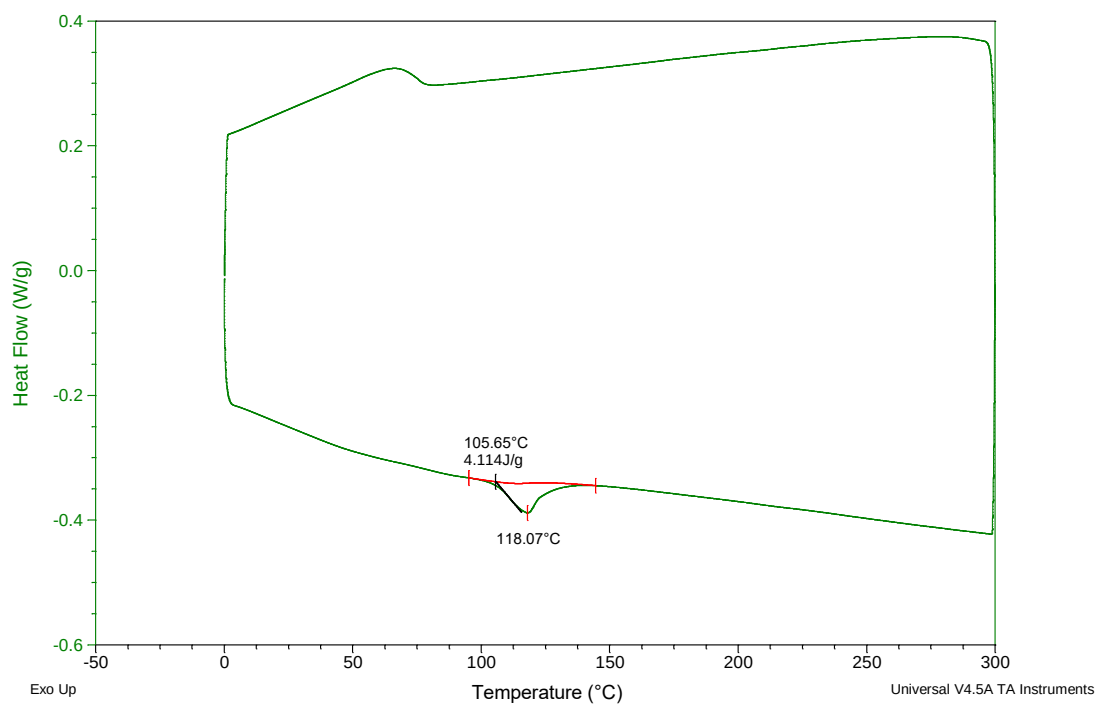
File: C:\...\C1C6C8-TEG-DPP-C5.sav
Operator: ZKK
Run Date: 13-Nov-2015 21:04
Instrument: DSC Q2000 V24.11 Build 124



Sample: C1C6C8-DPP-C5
Size: 4.1000 mg
Method: DPP-Standard

DSC

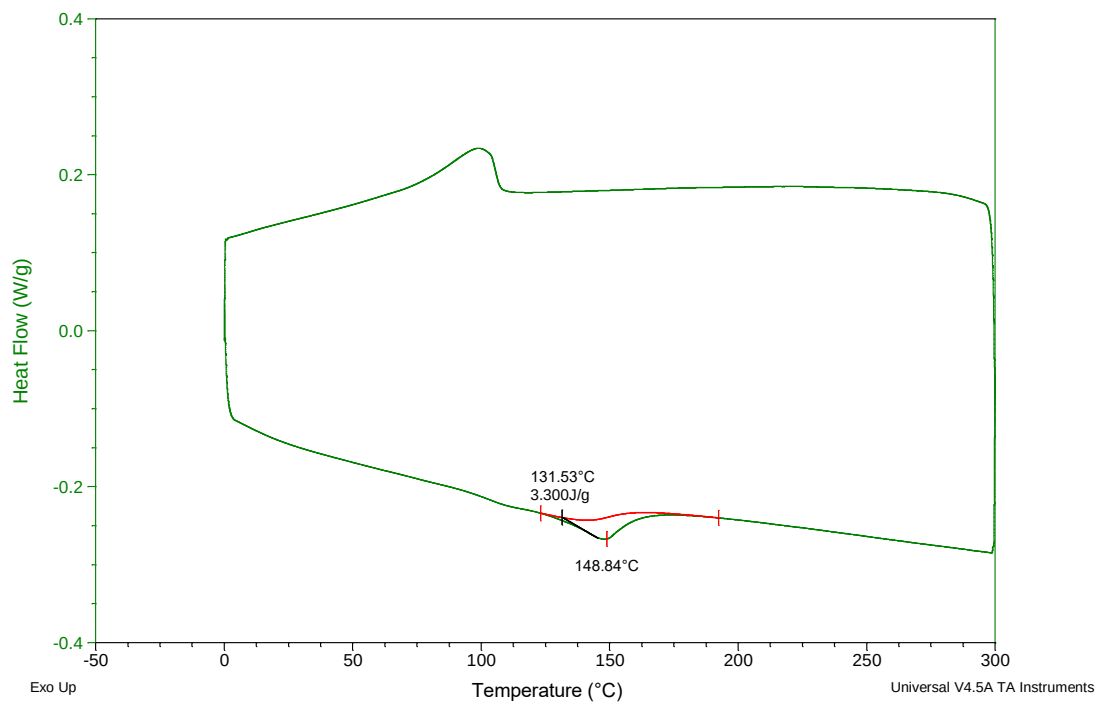
File: C:\...\C1C6C8-DPP-C5\C1C6C8-DPP-C5.sav
Operator: ZKK
Run Date: 13-Nov-2015 15:57
Instrument: DSC Q2000 V24.11 Build 124



Sample: TEG-DPP-C5
Size: 3.9000 mg
Method: DPP-Standard

DSC

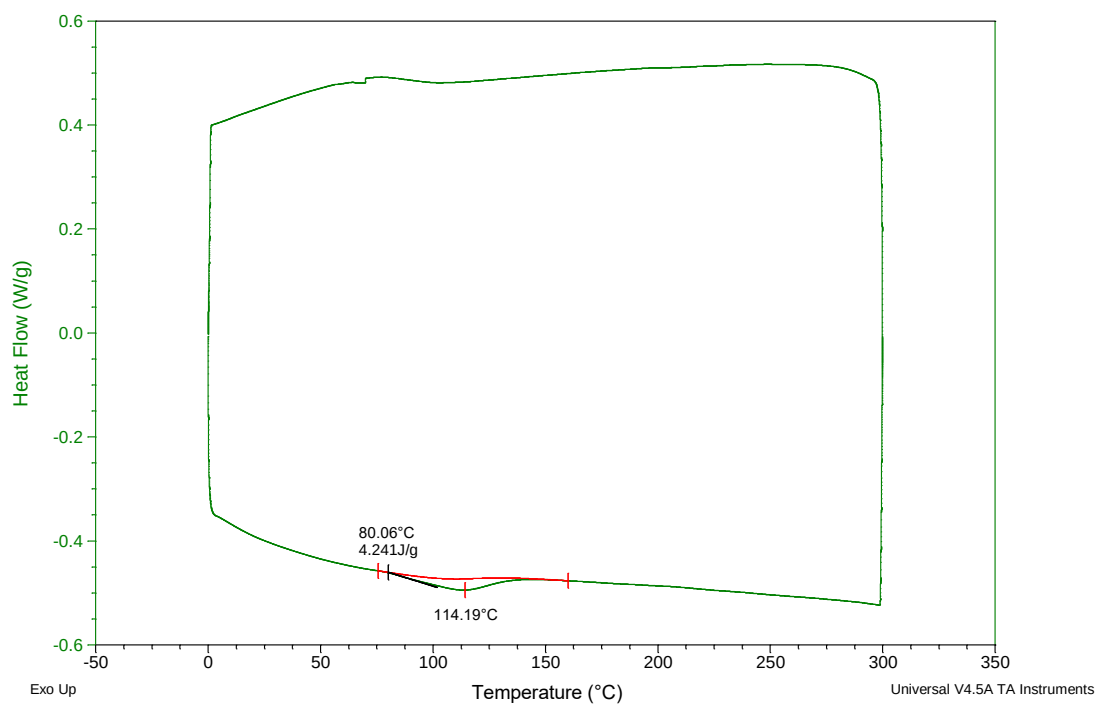
File: C:\...\TEG-DPP-C5\TEG-DPP-C5.sav
Operator: ZXX
Run Date: 13-Nov-2015 18:12
Instrument: DSC Q2000 V24.11 Build 124



Sample: Si-C1C6C8-DPP-C5
Size: 2.5000 mg
Method: DPP-Standard

DSC

File: C:\...\Si-C1C6C8-DPP-C5.sav
Operator: ZXX
Run Date: 10-Feb-2016 14:00
Instrument: DSC Q2000 V24.11 Build 124



Sample: Si-DPP-C5
Size: 2.5000 mg
Method: DPP-Standard

DSC

File: C:\...\Si-DPP-C5\Si-DPP-C5.sav
Operator: ZKK
Run Date: 09-Feb-2016 12:58
Instrument: DSC Q2000 V24.11 Build 124

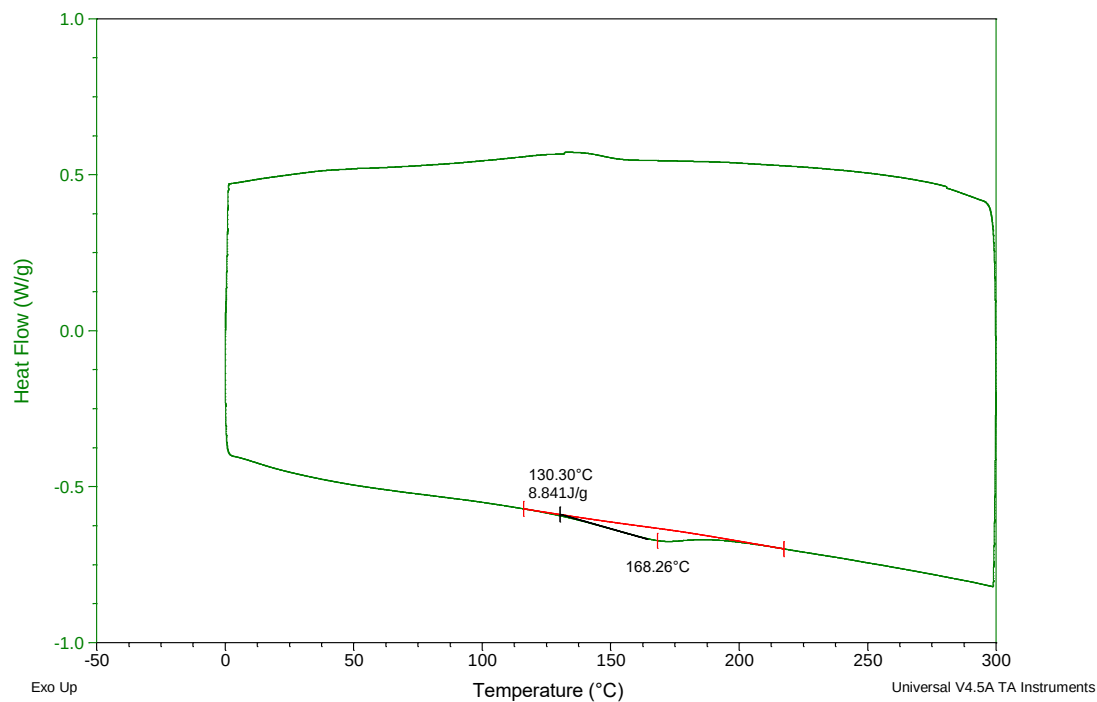


Figure S2 DSC thermograms of DPP polymers with different side chains. Exotherm up

Table S1. Mobilities of matrix polymers annealed at different temperatures

Polymer	Melting point °C	Processing method	Average mobility cm^2/Vs at different annealing temperatures			
			80 °C	120 °C	160 °C	180 °C
C1C6C8-TEG-DPP-C5	105	10mg/mL, spin coating, 1500r.p.m.	5.4E-4	3.6E-4		
Si-C1C6C8-DPP-C5	114	10mg/mL, spin coating, 1500r.p.m.	2.5E-3	2.3E-3		
C1C6C8-DPP-C5	118	10mg/mL, spin coating, 1500r.p.m.	2.3E-3	1.4E-3		
C3C10C10-DPP-C5	131	10mg/mL, spin coating, 1500r.p.m.	3.7E-3	4.2E-3	3.6E-3	
TEG-DPP-C5	149	5mg/mL, spin coating, 1500r.p.m.	8.6E-5	1.9E-4	1.0E-5	
Si-DPP-C5	168	5mg/mL, spin coating, 1500r.p.m.	2.1E-3	2.5E-3	2.8E-3	2.0E-3

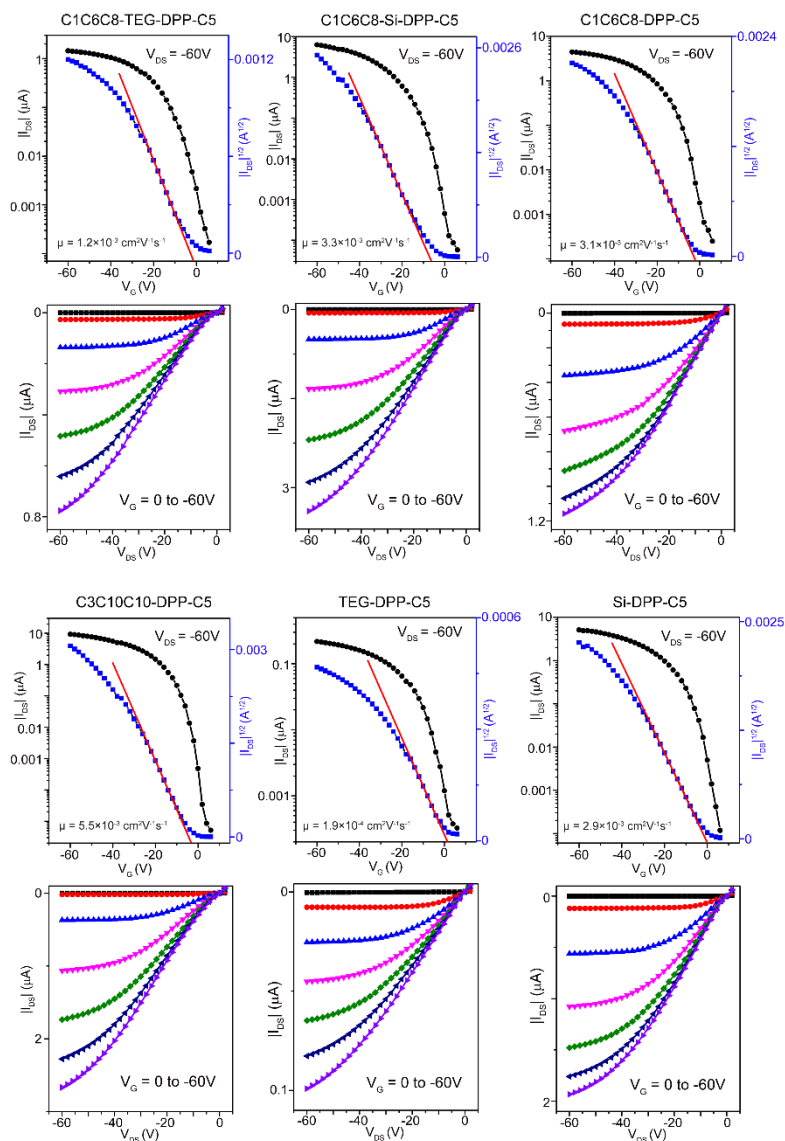


Figure S3. Typical output and transfer curves of the pure polymers.

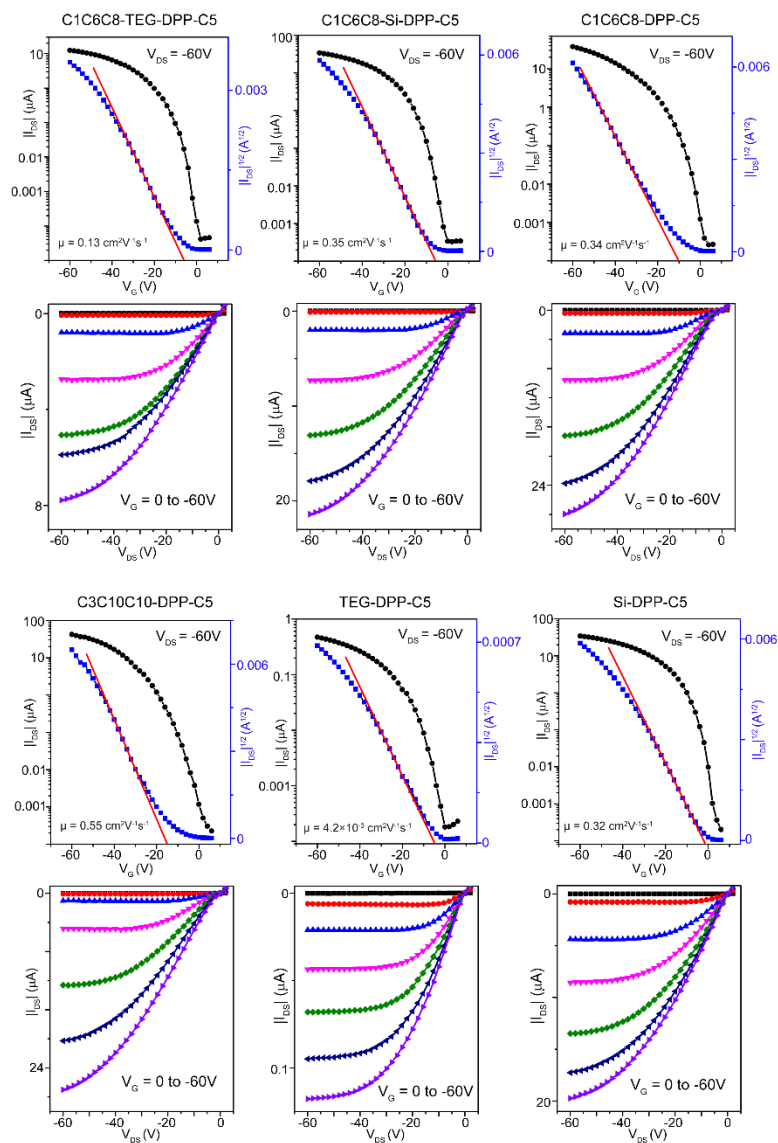


Figure S4. Typical output and transfer curves of the *c*-SPBs with 5 wt% of DPP-C0.

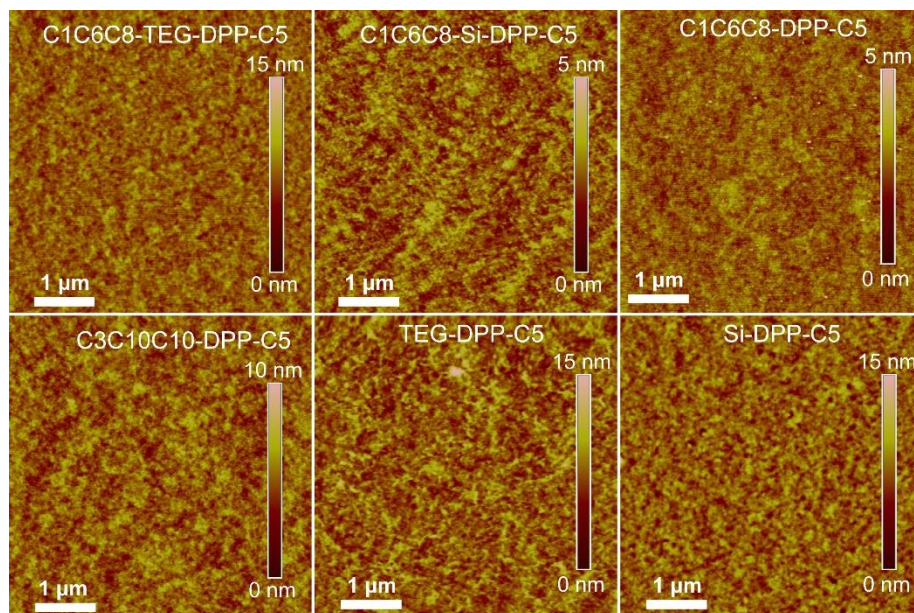


Figure S5. The AFM images of pure polymer films. The images were recorded as cast.

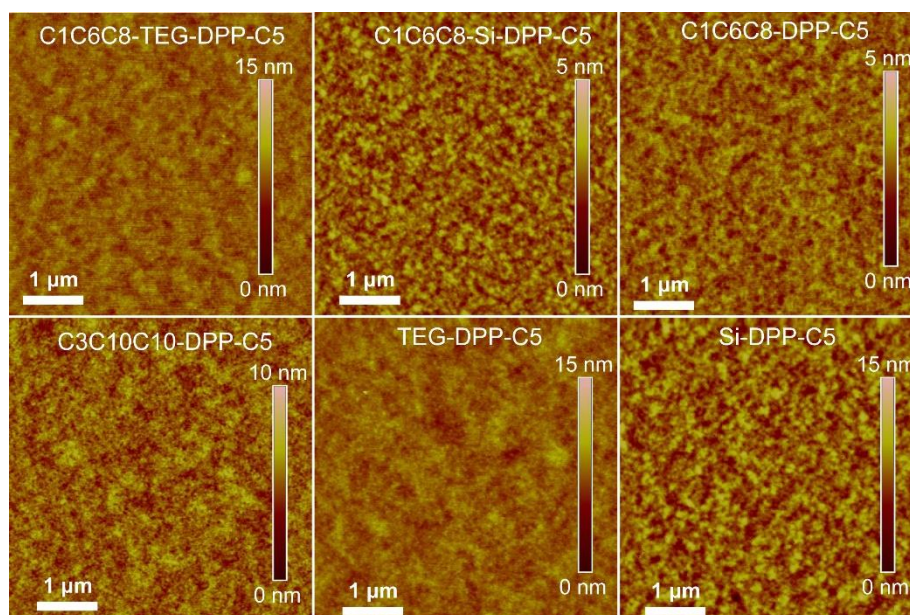


Figure S6. The AFM images of pure polymer films after annealed.

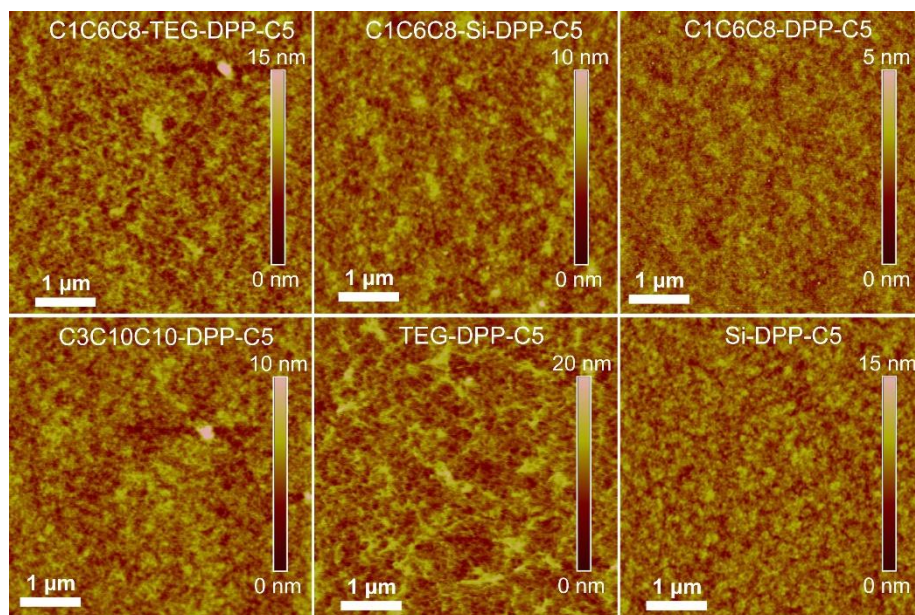


Figure S7. The AFM images of *c*-SPB films with 5 wt% of DPP-C0. The images were recorded as cast.

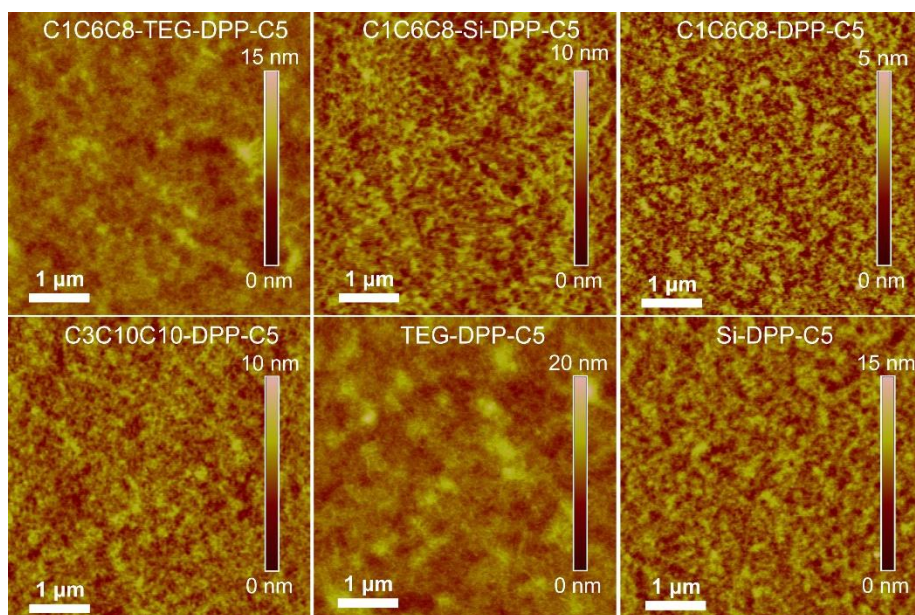


Figure S8 The AFM images of *c*-SPB films with 5 wt% of DPP-C0 after annealed.

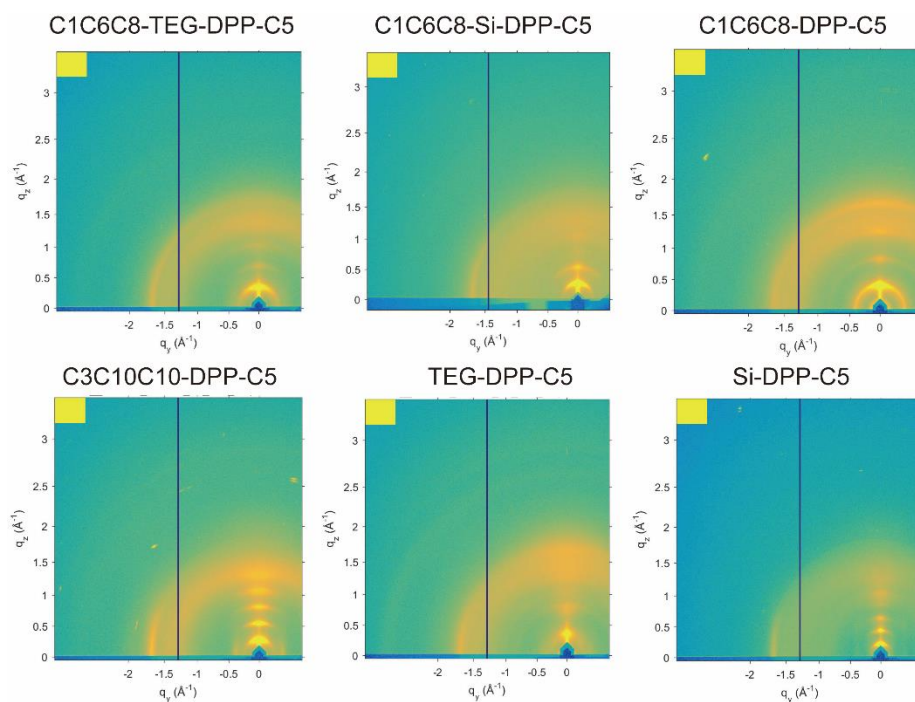


Figure S9 GIXRD images for pure matrix polymer with different side chains.

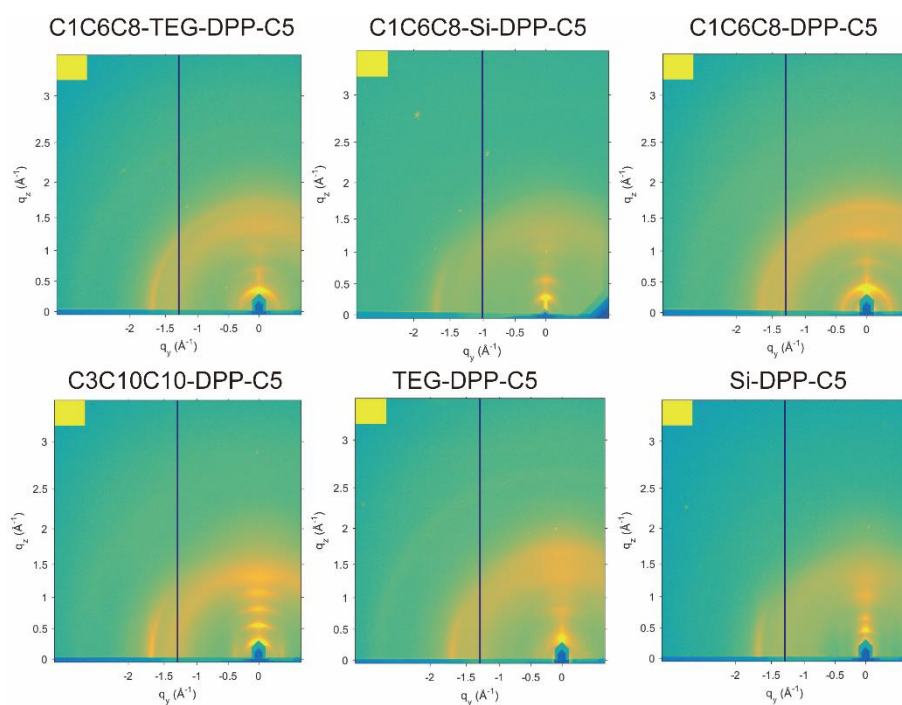


Figure S10 GIXRD images for c-SPBs with 5 wt% C3C10C10-DPP-C0.

UPS measurement: UPS measurements were performed in a PHI 5600 UHV system containing an 11-inch diameter hemispherical electron energy analyzer with a multichannel plate detector. UPS measurements were performed using the Excitech H Lyman- α photon source (E-LUX™121) coupled with a 90° ellipsoidal mirror (E-LUX™ EEM Optical Module). Samples were negatively biased (−5 V) during UPS measurements and the pass energy was 5 eV. With the exception of the beam path environment experiments, all UPS measurements were carried out with a dry house nitrogen purge of the beam path at 8 Torr. For details, please refer to Boehm, A. M.; Wieser, J.; Butrouna, K.; Graham, K. R. *Org. Electron.* 2017, 41, 9.

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