

Supporting Information

Tetracyanocyclopentadienide (TCCp)-Based Stable Poly(aromatic) Anions

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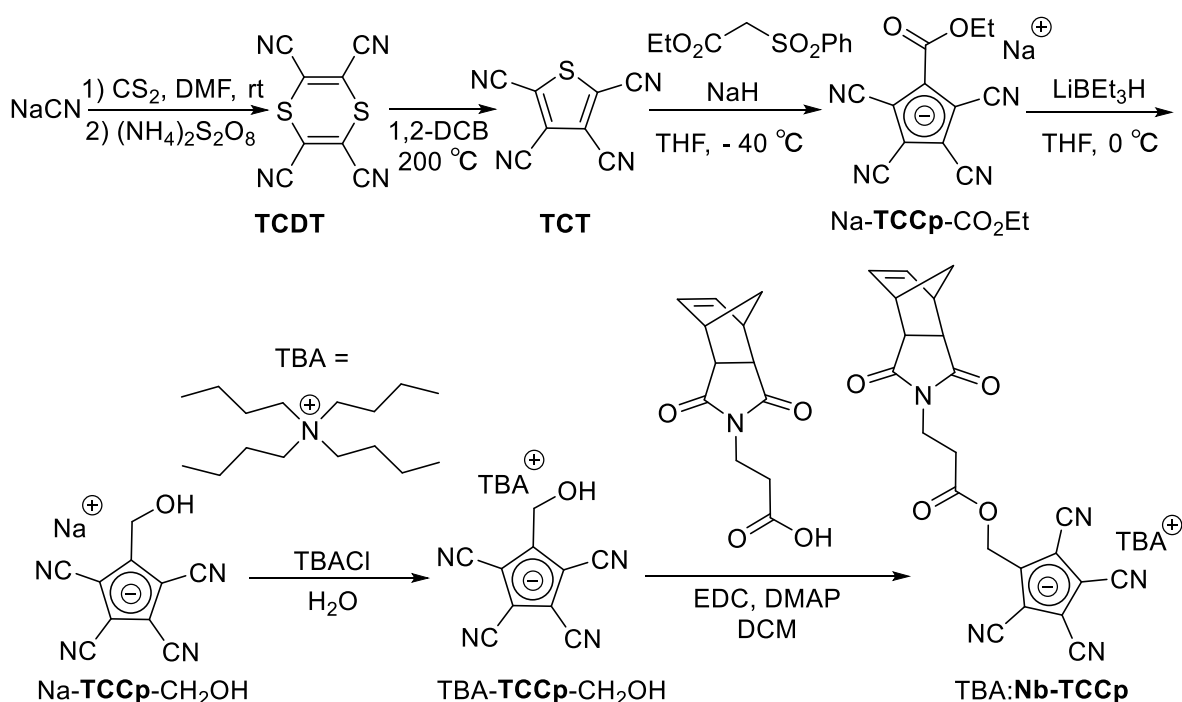
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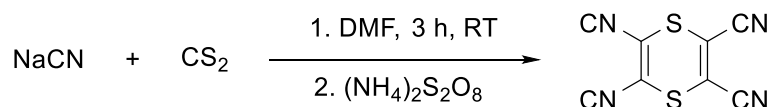
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General Considerations

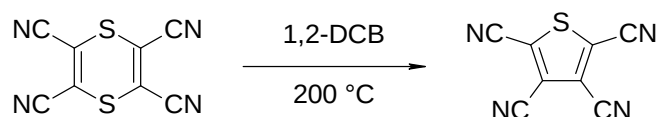
All reactions were performed under nitrogen atmosphere in oven-dried glassware following standard Schlenk techniques. Anhydrous solvents were purchased from Sigma-Aldrich and used without further purification. Automated preparative chromatography was performed with a Yamazen Smart Flash AI-580S-Premiun. ^1H NMR and ^{13}C NMR were recorded on Bruker AV-III-400 and Bruker Avance-III-800 instruments with chemical shifts reported relative to residual solvent. Infrared spectra were measured with a Thermo Nicolet Nexus FT-IR. HRMS data were obtained on an Thermo LTQ-Orbitrap mass spectrometer with a FTMS analyzer. Glass transition temperature, T_g , was measured using a TA Instruments DSC Q 2000. Sample dispersity weights were obtained using a TOSOH GPC with DMF (with 0.01 M LiCl) vs. polystyrene standards at 50 °C. Polymer molecular weights (M_n) were determined using end group analysis via ^1H NMR on the Bruker Avance-III-800 MHz NMR. UV-vis was measured using an integrated sphere detector from (400 – 1100 nm). Atomic force microscopy (AFM) images were collected on a Veeco Dimension 3100 atomic force microscope.

General Procedure A (Synthesis of TBA:Nb-TCCp Monomer)

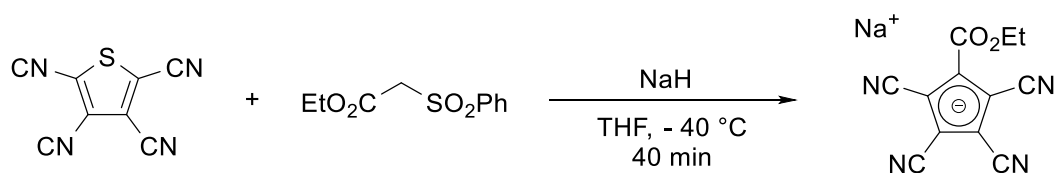




Tetracyanodithiin 1 (TCDT) was prepared following literature procedure.¹ To a suspension of NaCN (23.5 g, 1.0 eq) in DMF (260 mL), CS₂ (28.8 mL, 1.0 eq) was added dropwise via a pressure equalizing dropping funnel. The reaction was stirred at room temperature for 3 h. The reaction was then precipitated into water (1400 mL) and was left undisturbed for 14 h. The precipitated solids were removed by filtration, and the mother liquor was collected into a beaker. To the reaction in the beaker, a solution of ammonium sulfate (109.5 g, 1.0 eq in 210 mL of water) was added dropwise over 40 min. Following addition, the reaction mixture was stirred for an additional 30 min. The resulting solids were collected by filtration, washed with water, and dried. The solids were suspended in MeCN (1400 mL), stirred for 30 min, then filtered. The filtrate was collected and concentrated via rot evap to yield tetracyanodithiin (17.5 g, brown/orange solid) which was used directly in the next step.

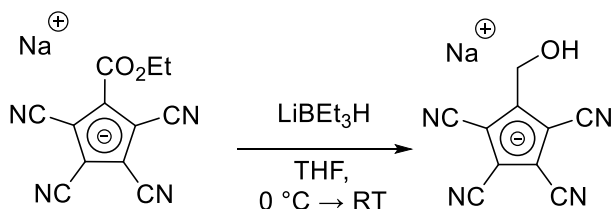


Tetracyanothiophene 2 (TCT) was synthesized following literature procedure.¹ TCDT (6.5 g) was dissolved into 1,2-dichlorobenzene (80 mL) into a 100-mL Schlenk tube. The reaction mixture was degassed by three freeze-pump-thaw cycles via a liquid nitrogen bath. The reaction was then stirred for 2 h at 200 °C. Once cooled, the reaction was poured into hexanes (300 mL) and mixed. The resulting brown solids were collected via filtration. The solids were dissolved into EtOAc (300 mL) and passed through a short plug of silica. The filtered solution was then concentrated via rot evap. The resulting dark brown solid was dissolved in the minimum amount of EtOAc and precipitated into hexanes (300 mL). The resulting brown solids were collected and dried (5.5 g, 83%).

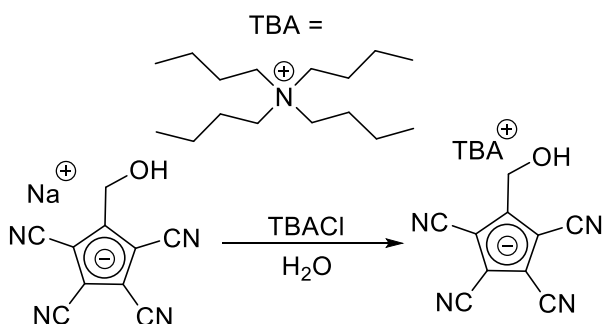


Sodium 1,2,3,4-Tetracyano-5- (ethoxycarbonyl)cyclopentadienide 3 (Na-TCCp-CO₂Et) was synthesized following literature procedure.¹ A suspension of NaH (60% dispersed in mineral oil, 2.6 g, 3.0 eq) in THF (28 mL) was cooled to 0 °C. Once at temperature, a solution of ethyl 2-(phenylsulfonyl)acetate (5.0 g, 1.0 eq in 58 mL THF) was added dropwise. The reaction mixture was stirred at 0 °C for 30 min. The reaction was then cooled to -40 °C. After cooling, a solution of TCT (4.4 g, 1.1 eq) in THF (58 mL) was added dropwise. The reaction was stirred at -40 °C for 40 min. After 40 min, the reaction was quenched with 80 mL of saturated NaCl solution and warmed to RT. The resulting

reaction mixture was extracted with EtOAc. The organic phase was washed with brine, dried with Na₂SO₄, and concentrated via rot evap. The reaction was then passed through a short silica plug (90/10 EtOAc/MeCN). The collected reaction was then concentrated. The dark brown solid was then dissolved in the minimum amount of MeCN and precipitated into toluene (300 mL). The resulting precipitates were collected by filtration and dried as a light brown solid (5.0 g, 88%).



Sodium 1,2,3,4-Tetracyano-5-(hydroxymethyl)-cyclopentadienide 4 (Na-TCCp-CH₂-OH) was prepared following literature procedure.¹ Na-TCCp-CO₂Et (4.0 g, 1.0 eq) was dissolved into THF (80 mL). The reaction was chilled to 0 °C. To the chilled reaction, LiEt₃H (1.0 M, 42.3 mL, 2.75 eq) was added dropwise. After addition, the reaction was warmed to RT and stirred for 2 h. The reaction was quenched with NaHCO₃ (40 mL) and a 10% solution of Rochelle's salt (in water, 80 mL) and stirred for several hours. The reaction was then extracted with EtOAc. The organic phase was washed with brine, dried with Na₂SO₄, and concentrated via rot evap. The reaction was purified by column chromatography (10 → 20% MeOH/CH₂Cl₂) as an off-white/beige solid (2.68g, 81%).



Tetrabutylammonium 1,2,3,4-Tetracyano-5-(hydroxymethyl)-cyclopentadienide 5 (TBA-TCCp-CH₂-OH) was prepared by mixing a solution of Na-TCCp-CH₂-OH (2.5 g, 1.0 eq) in water with a solution of tetrabutylammonium chloride (3.18 g, 1.0 eq) in water. Upon addition of the two solutions, orange solids precipitated. The solids were collected by filtration, washed with water, and dried thoroughly to yield a light orange solid (97% yield). ¹H NMR (400 MHz, CD₃CN): δ ppm 4.48 (d, J = 5.7 Hz, 2H), 3.14 (t, J = 5.7 Hz, 1H), 3.08 – 3.04 (m, 8H), 1.62 – 1.54 (m, 8H), 1.33 (sex, J = 7.4 Hz, 8H), 0.95 (t, J = 7.2 Hz, 12H); ¹³C NMR (MHz, CD₃CN): δ ppm 139.1, 116.5, 116.1, 100.4, 97.2, 59.3, 57.1, 24.2, 20.3, 13.8; IR (neat, cm⁻¹): 3426, 2961, 2204, 1375. HRMS m/z calcd for C₁₆H₃₆N⁺ [M+H]⁺ 242.2838, obsd 242.28423; m/z calcd for C₁₀H₃ON₄⁺ [M+H]⁺ 195.03123, obsd 195.03146

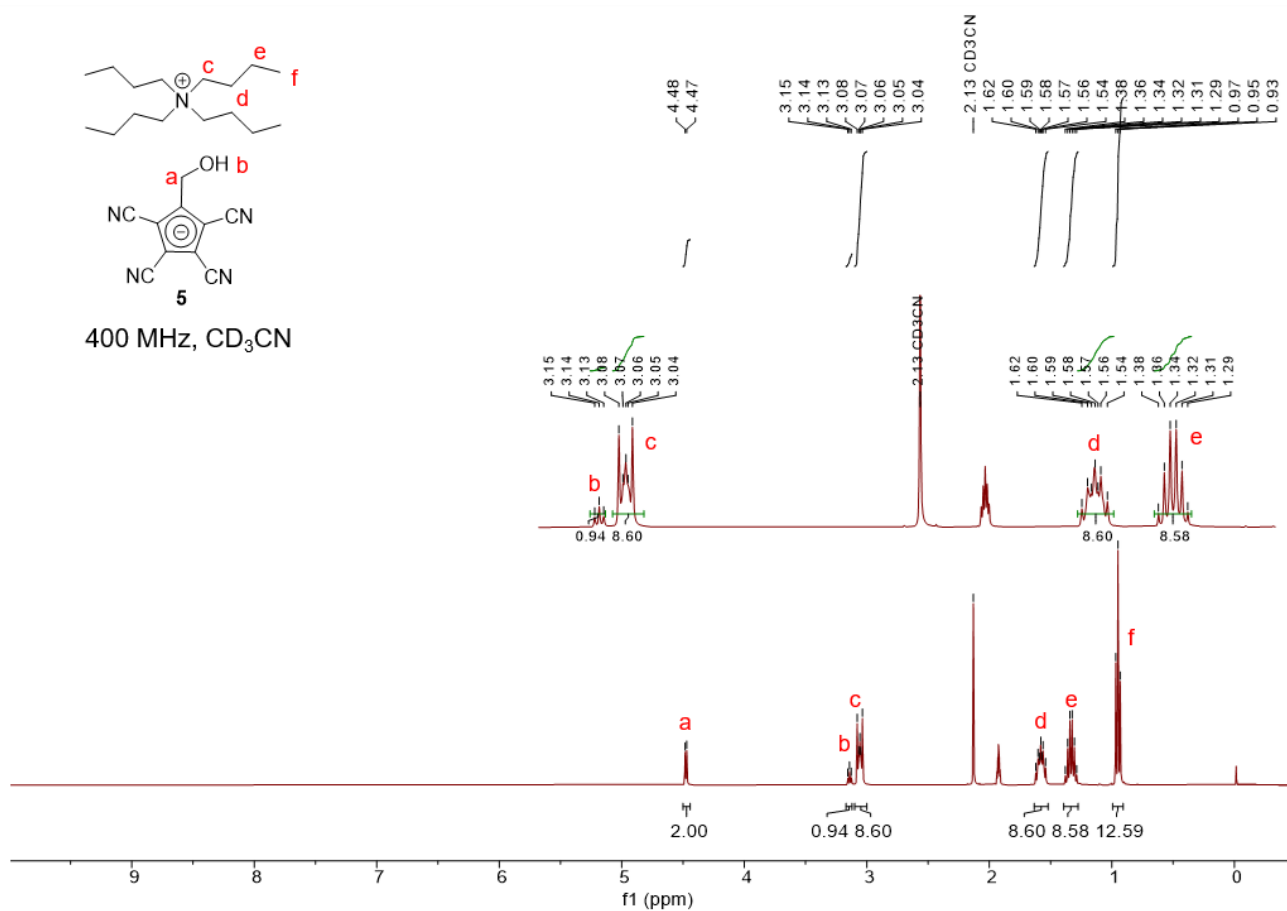


Fig S1. ¹H NMR of TBA-TCCp-CH₂-OH 5.

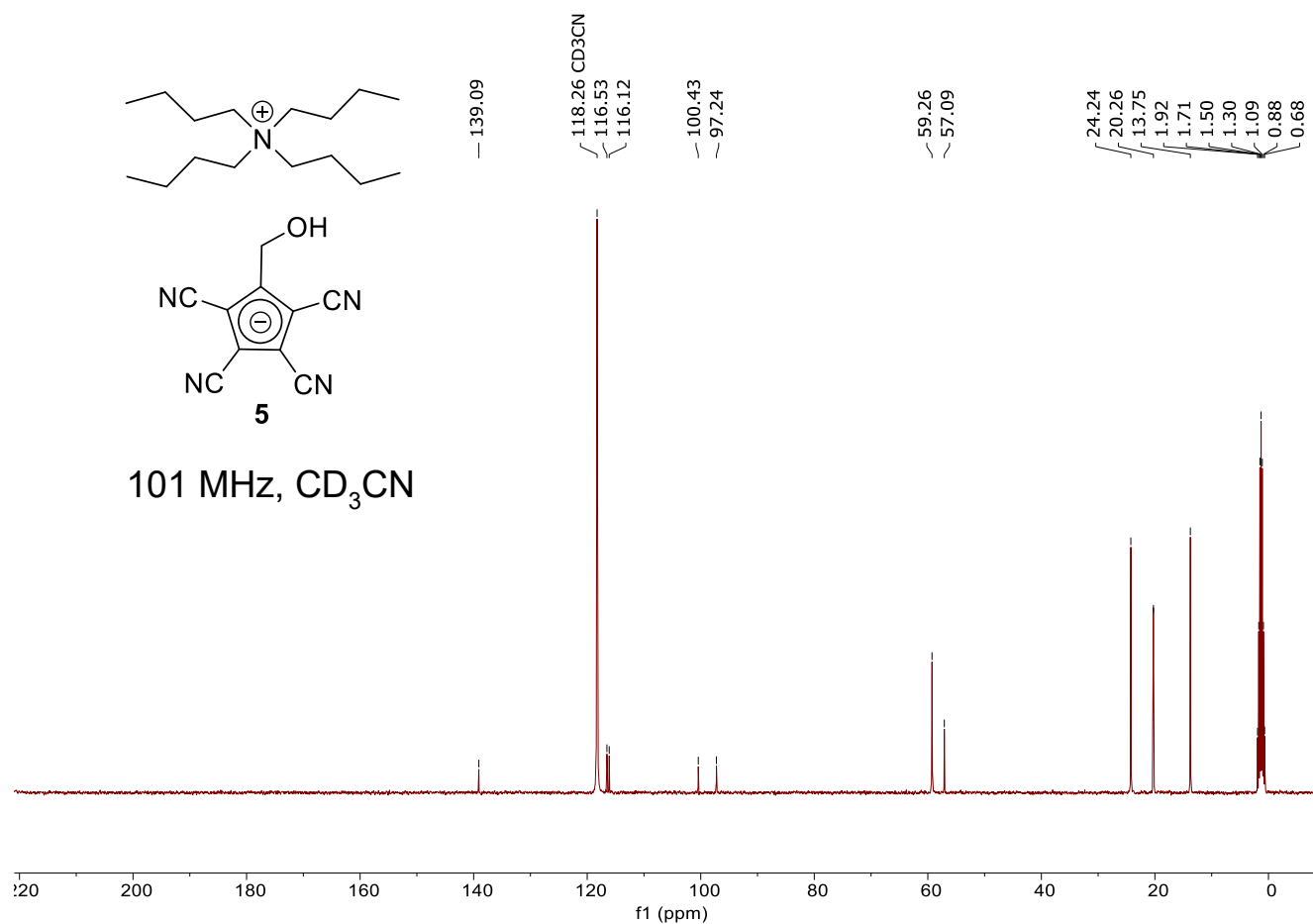
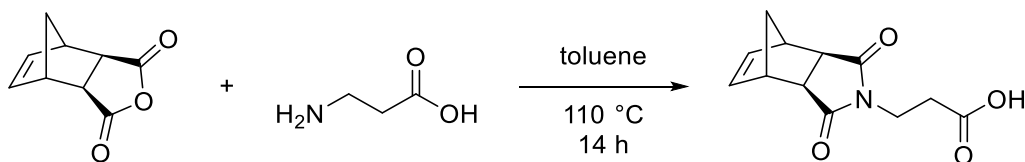
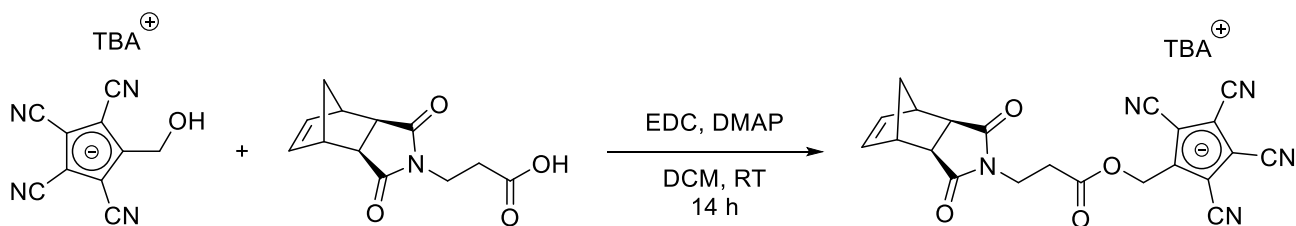


Fig S2. ¹³C NMR of TBA-TCCp-CH₂-OH **5**.



Norbornene carboxylic acid imide 6 was prepared following literature procedure.² Norbornene exo-anhydride (4.0 g, 1.0 eq), 3-aminopropanoic acid (3.2 g, 1.0 eq), and toluene (26 mL) were mixed together in a round bottom flask equipped with a reflux condenser. The reaction was heated to reflux (110 °C) for 16 h, then cooled to room temperature. The toluene was removed via rotary evaporation until dryness. The crude solid was dissolved into dichloromethane, washed with water, then dried with sodium sulfate. The solvent was stripped in vacuo to yield a white solid with no further purification (6.2 g, 93%).



TBA:Nb-TCCp 7 was prepared via Steglich Esterification protocol. TBA-TCCp-CH₂-OH (2.0 g, 1.0 eq), Norbornene carboxylic acid imide (1.08 g, 1.0 eq), 1-Ethyl-3-(3-dimethylaminopropyl)carbodiimide (2.48 g, 3.5 eq), 4-dimethylaminopyridine (0.11 g, 0.2 eq), and dichloromethane (46 mL) were mixed into a 250 mL round bottom flask. The reaction was stirred for 14 h at room temperature. The reaction was washed with 1 M HCl and extracted with dichloromethane. The resulting organic phase was mixed with a solution of tetrabutylammonium (1.3 g, 15 mL in water) for 15 min. The organic layer was separated, dried with Na₂SO₄, and passed through celite. The filtered solution was washed with 1M NaHCO₃, dried with Na₂SO₄, and the solvent was removed in vacuo. The crude solid was purified by automated preparative column chromatography (95/5 DCM/MeOH → 80/20 DCM/MeOH) to yield a light orange/beige solid (2.1 g, 70%). ¹H NMR (400 MHz, CD₃CN): δ ppm 6.26 (t, J = 1.8 Hz, 2H), 5.0 (s, 2H), 3.64 (t, J = 7.1 Hz, 2H), 3.09 – 3.03 (m, 10H), 2.58 – 2.55 (m, 4H), 1.61 – 1.53 (m, 9H), 1.37 – 1.28 (m, 10H), 1.19 (d, J = 9.8 Hz, 1H), 0.94 (t, J = 7.3 Hz, 12H); ¹³C NMR (MHz, CDCl₃): δ ppm 177.3, 170.0, 137.5, 130.9, 115.2, 115.1, 100.1, 97.2, 58.6, 47.5, 44.9, 42.5, 34.0, 31.6, 23.5, 19.4, 13.3; IR (neat, cm⁻¹): 2963, 2676, 2206, 1694; HRMS m/z calcd for C₁₆H₃₆N⁺[M+H]⁺ 242.28423, obsd 242.28345; m/z calcd for C₂₂H₁₄O₄N₅⁺ [M+H]⁺ 412.10573, obsd 412.10368.



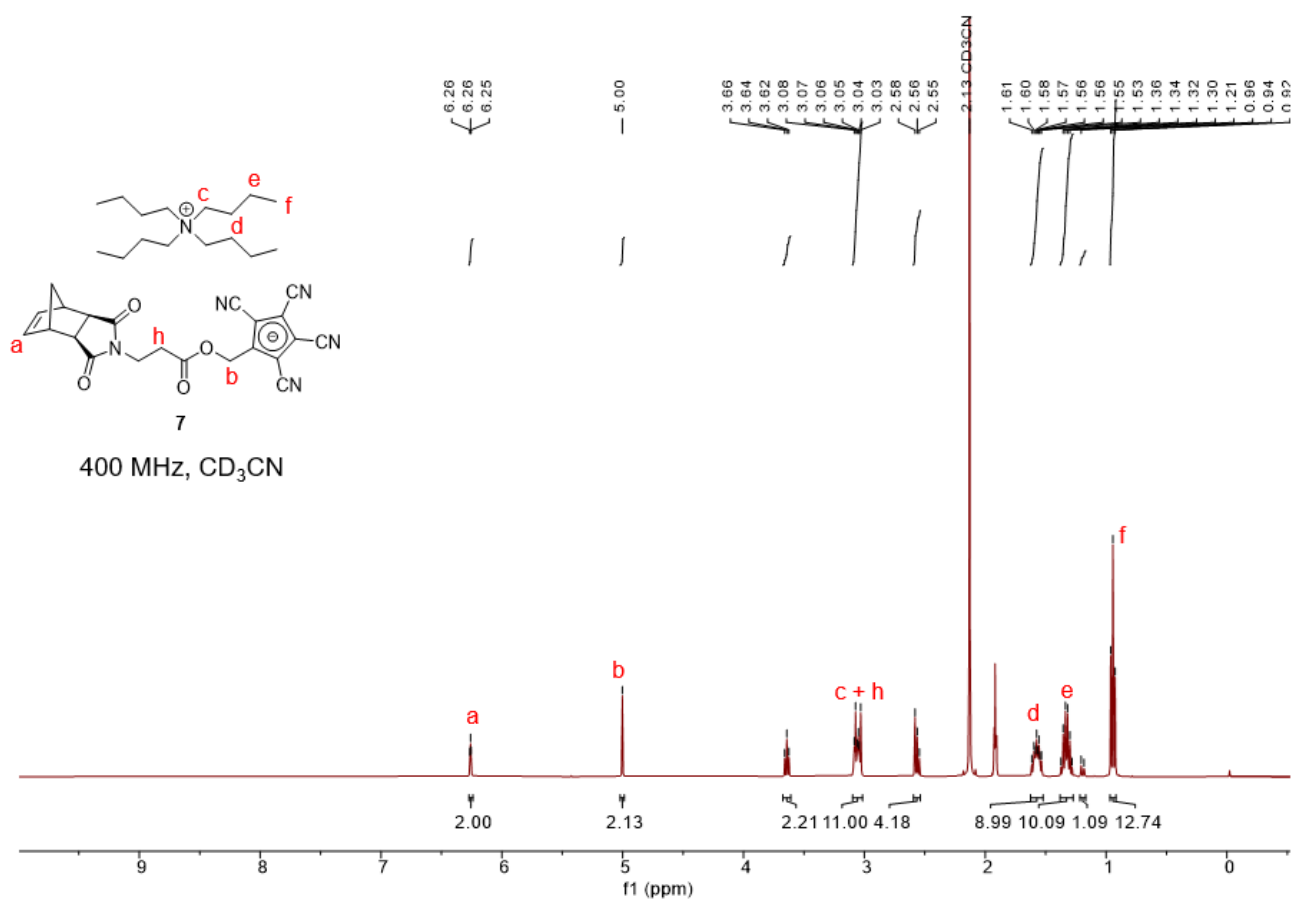


Fig S3. ¹H NMR of TBA:Nb-TCCp 7.

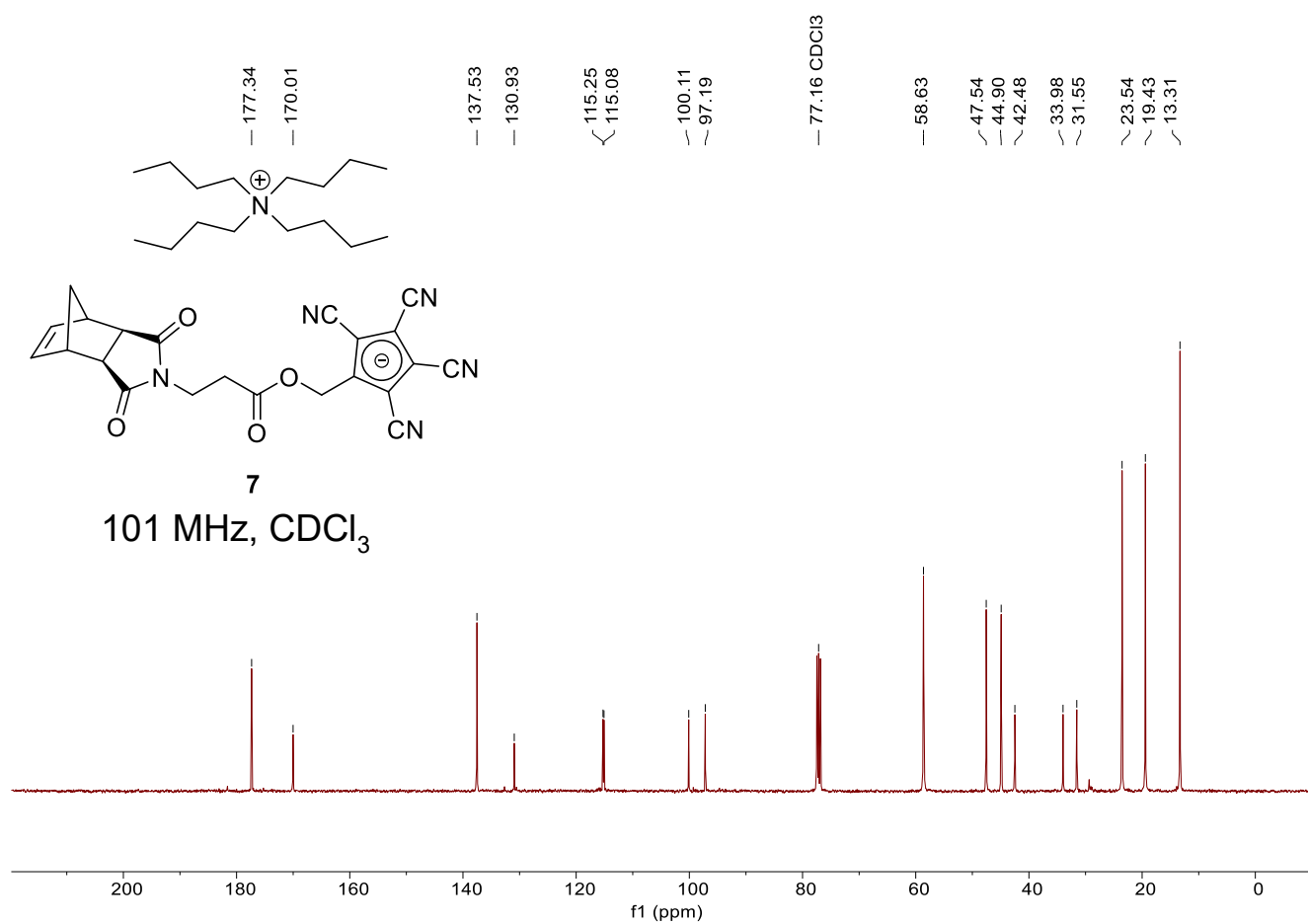


Fig S4. ¹³C NMR of TBA:Nb-TCCp **7**.

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Chemical reaction scheme showing the reaction of a ruthenium complex with pyridine. The reactant is a ruthenium complex with a chiral ligand (1,2-bis(mesityl)-4,5-dihydroimidazole), a phenyl group, and three chlorine atoms. The reaction conditions are RT, 5 min. The product is a ruthenium complex where one chlorine atom has been replaced by a pyridine ring.

Chemical reaction scheme showing the synthesis of a polymer-bound dendritic molecule. The starting material is a bicyclic imide with a side chain containing a tert-butyl ammonium cation (TBA⁺) and a 1,3,5-tricyanobenzene ring. This reacts with G3 in a solvent at room temperature (RT) for 2 hours to form a polymer-bound product where the side chain is attached to a polymer backbone via an ester linkage.



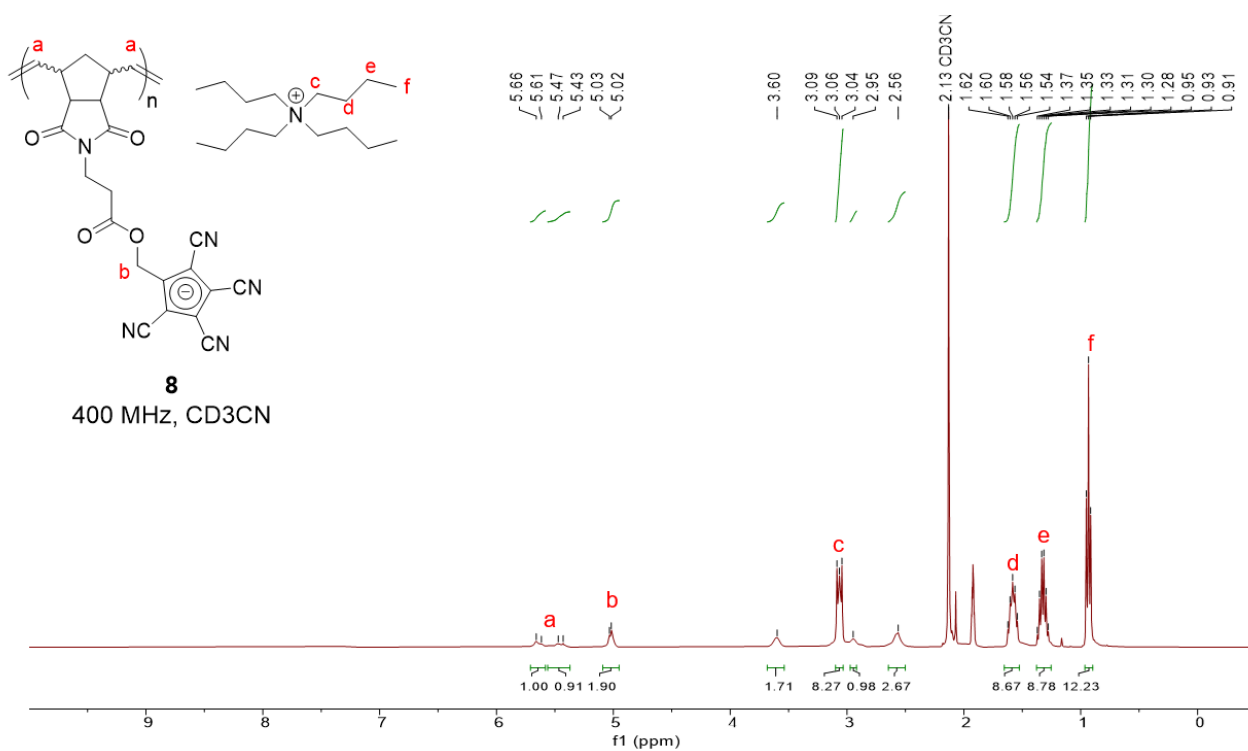


Fig S5. ^1H NMR of TBA:PNb-TCCp **8**.

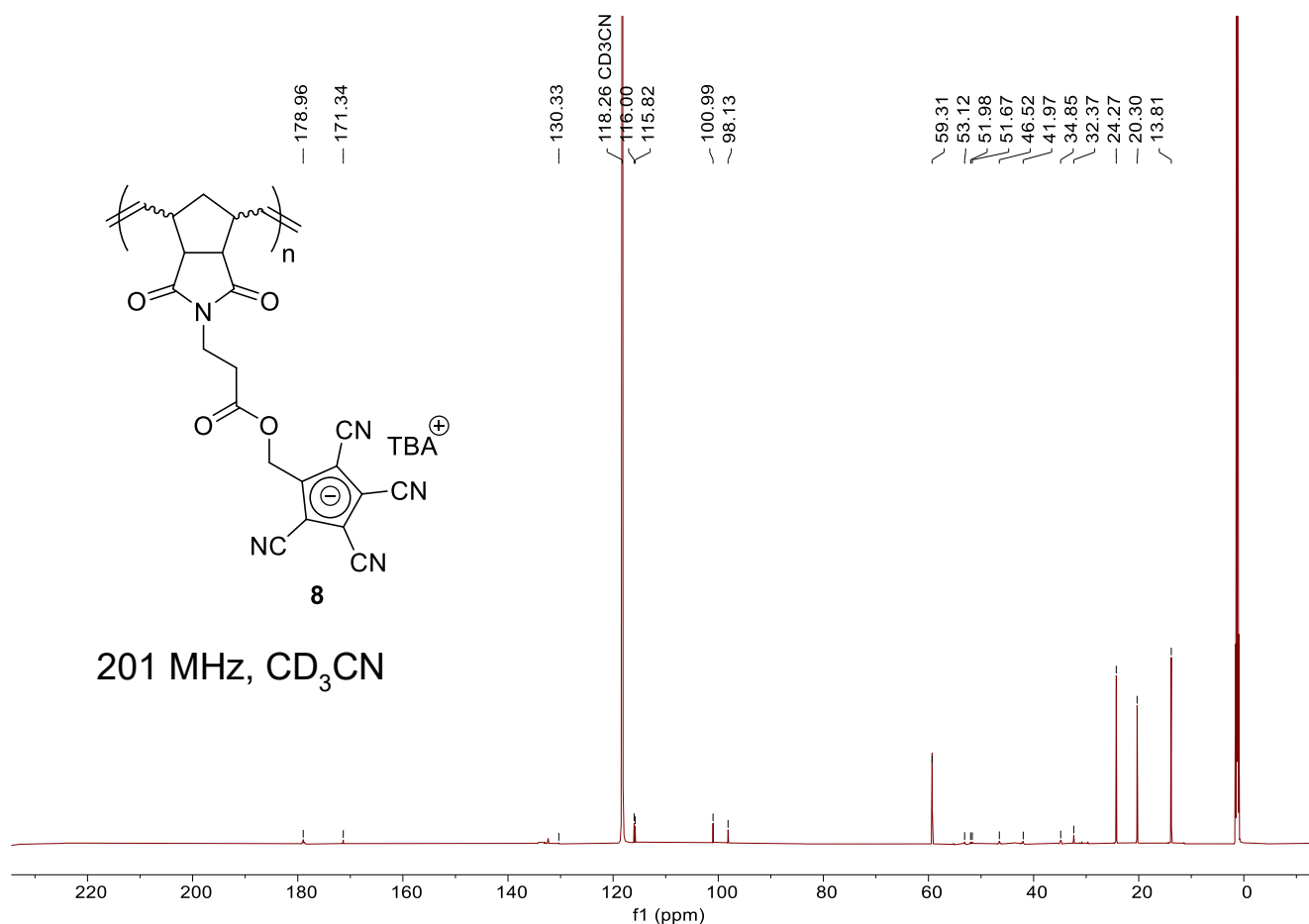


Fig S6. ¹³C NMR of TBA:PNb-TCCp **8**.

Table S1. Solubility chart of monomer **7** and polymer **8**.

	Monomer 7	Polymer 8
Soluble	DCM, chloroform Ethyl acetate, DMF, Acetone, MeCN, MeOH	Acetone, MeCN, DMF
Insoluble		DCM, chloroform, THF, Ethyl acetate, MeOH

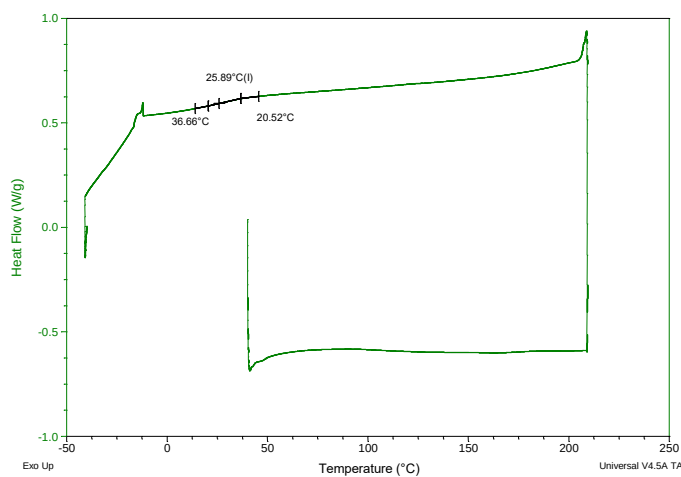


Fig S7. DSC thermogram of TBA:PNb-TCCp **8**, 1st heat, exo up.

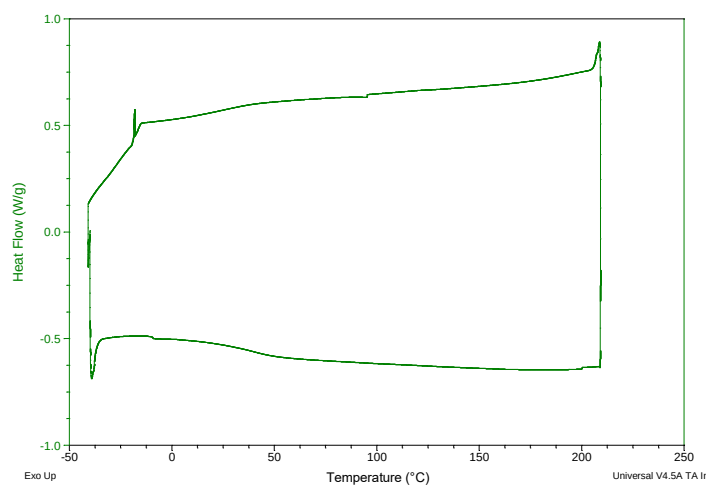


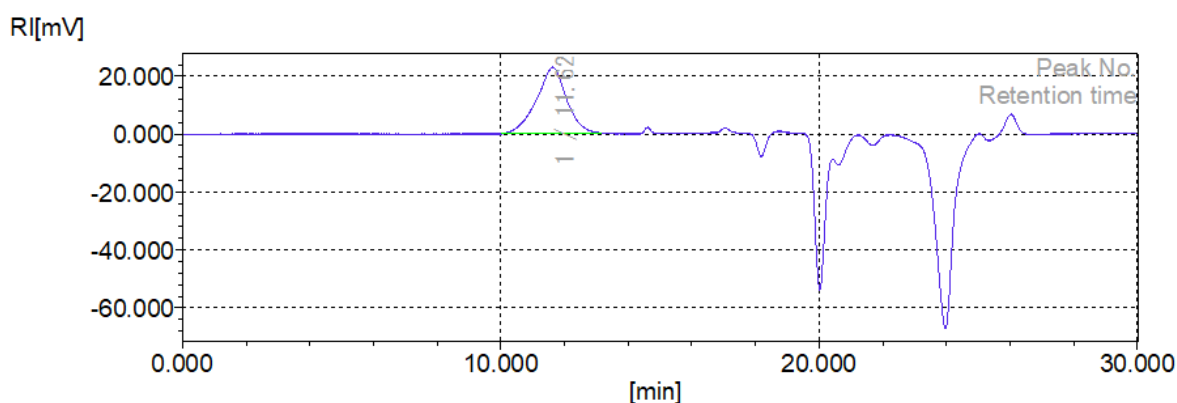
Fig S8. DSC thermogram of TBA:PNb-TCCp **8**, 2nd heat, exo up.

Note: Sample dispersity weights were obtained using a TOSOH GPC with DMF (with 0.01 M LiCl) vs. polystyrene standards at 50 °C. Using a polystyrene calibration standard returned M_n values that are much larger than the theoretical estimates. End group analysis of the resulting polymers was performed using ^1H NMR obtained from Bruker Avance-III-800 MHz NMR. This returned more precise estimates and were reported as noted. The raw GPC chromatographs using the polystyrene standards are shown below.

Table S2. ROMP of TBA:Nb-TCCp Using **G3**, Solvents Effect on the Degrees of Polymerization and Polydispersities

Entry	Solvent	Yield (%)	M_n , Theo (kDa)	M_n , NMR (kDa) ^b	$\bar{D}$ ^c
1	DCM	98	16.4	10.0	1.28
2	THF	95	16.4	12.7	1.15
3	DMF	95	16.4	11.9	1.20
4^a	DMF	99	32.7	23.6	1.43

^aPerformed at rt for 2 h using 4 mol% **G3** catalyst on 1.2 mmol scale under N_2 ; [monomer **7**] = 0.2 M; isolated yields. ^bDetermined by end group analysis of ^1H NMR data. ^cDetermined by GPC. ^d2 mol% **G3** catalyst.

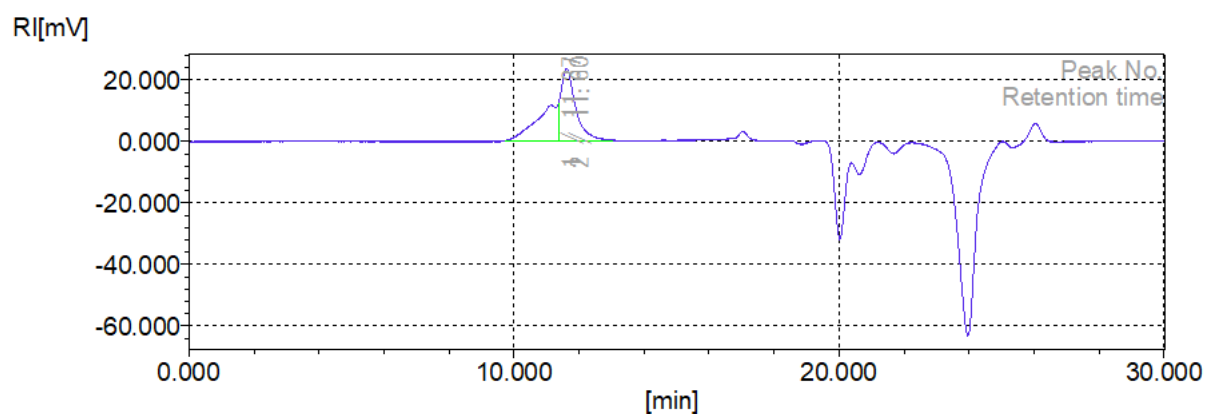


Result of molecular weight calculation (RI)

Peak 1 Base Peak

	[min]	[mV]	[mol]	M_n	122,620
Peak start	10.028	0.212	720,877	M_w	157,326
Peak top	11.627	23.198	132,558	M_z	202,371
Peak end	13.198	0.350	25,074	M_{z+1}	257,273
				M_v	157,326
Height [mV]	22.916			M_p	132,559
Area [mV*sec]	1481.420			M_z/M_w	1.29
Area% [%]	100.000			M_w/M_n	1.28
[eta]	157325.71388			M_{z+1}/M_w	1.63

Fig S9. Raw GPC chromatogram of Table S2. Entry 1.

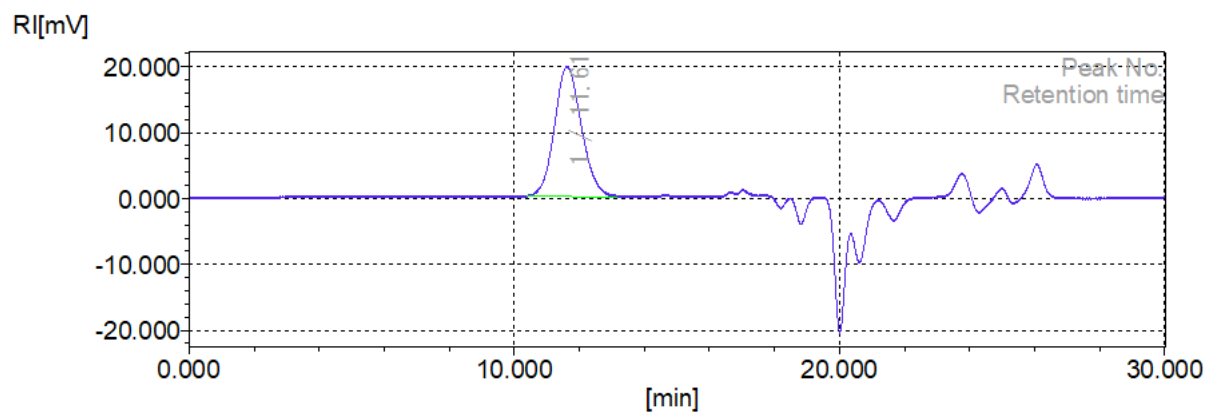


Result of molecular weight calculation (RI)

Peak 1 Base Peak

	[min]	[mV]	[mol]		
Peak start	9.725	0.357	994,114	Mn	274,840
Peak top	11.375	12.508	173,065	Mw	316,060
Peak end	11.375	12.508	173,065	Mz	373,454
				Mz+1	443,232
				Mv	316,060
Height [mV]	12.160			Mp	173,065
Area [mV*sec]	554.009			Mz/Mw	1.18
Area% [%]	43.191			Mw/Mn	1.15
[eta]	316060.15101			Mz+1/Mw	1.40

Fig S10. Raw GPC chromatogram of Table S2. Entry 2.

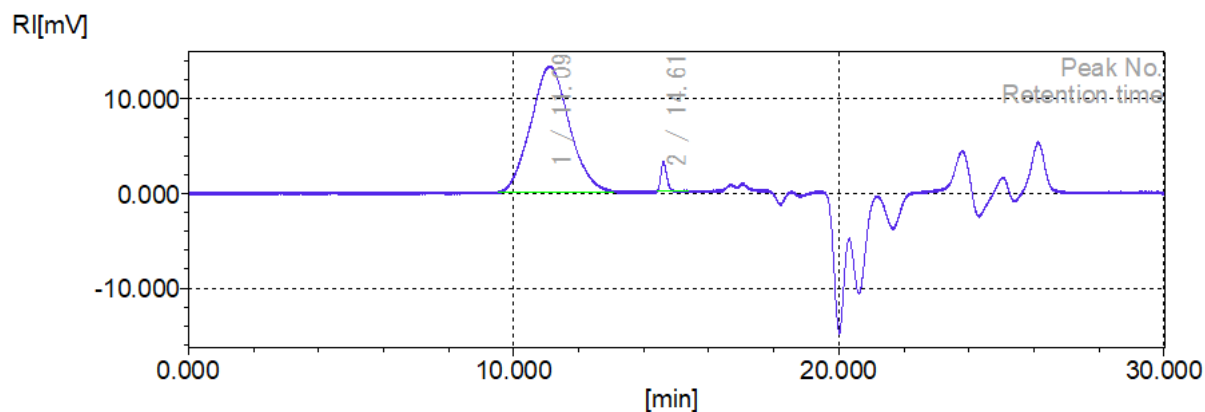


Result of molecular weight calculation (RI)

Peak 1 Base Peak

	[min]	[mV]	[mol]		
Peak start	10.377	0.389	498,402	Mn	114,316
Peak top	11.612	20.179	134,682	Mw	136,644
Peak end	13.198	0.226	25,074	Mz	160,781
				Mz+1	187,202
				Mv	136,644
Height [mV]	19.861			Mp	134,682
Area [mV*sec]	1134.075			Mz/Mw	1.18
Area% [%]	100.000			Mw/Mn	1.20
[eta]	136643.63797			Mz+1/Mw	1.37

Fig S11. Raw GPC chromatogram of Table S2. Entry 3.



Result of molecular weight calculation (RI)

Peak 1 Base Peak

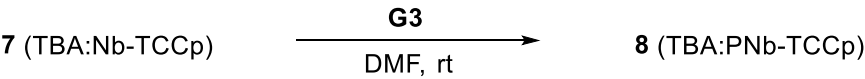
	[min]	[mV]	[mol]		
Peak start	9.508	0.107	1,250,641	Mn	182,389
Peak top	11.090	13.531	234,071	Mw	259,999
Peak end	13.112	0.169	27,485	Mz	353,682
				Mz+1	457,399
				Mv	259,999
Height [mV]	13.397			Mp	234,072
Area [mV*sec]	1096.788			Mz/Mw	1.36
Area% [%]	96.490			Mw/Mn	1.43
[eta]	259998.87738			Mz+1/Mw	1.76

Fig S12. Raw GPC chromatogram of Table S2. Entry 4.

General Procedure C (Catalyst Loading vs Yield and Molecular Weight)

General procedure follows polymerization of **8** with varying [monomer]/[initiator]:

Table S3. Controlled Polymerization Studies varying catalyst loading and concentrations.



Entry	[M]/[I]	Yield (%)	Concentration (mM)	M _n , NMR (kDa)	Đ
1	25	95	200	11.9	1.20
2	60	98	200	24.3	1.59
3	100	60	200	26.1	1.62
4	200	40	200	27.8	1.61

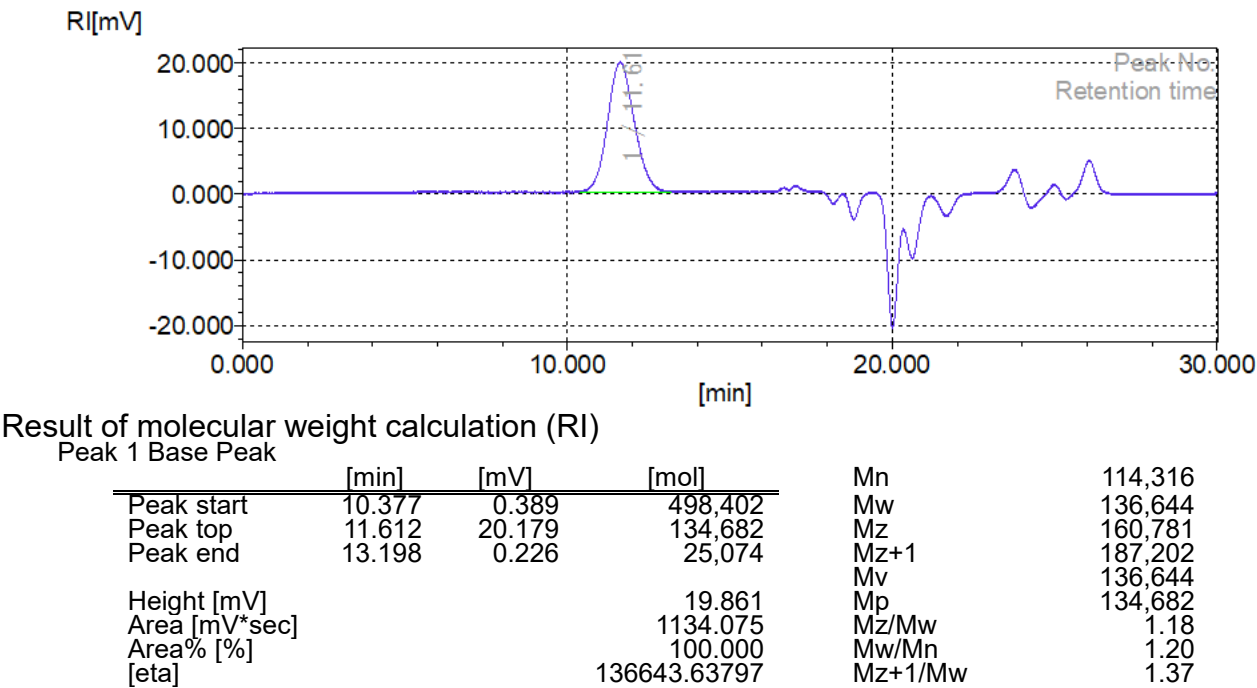
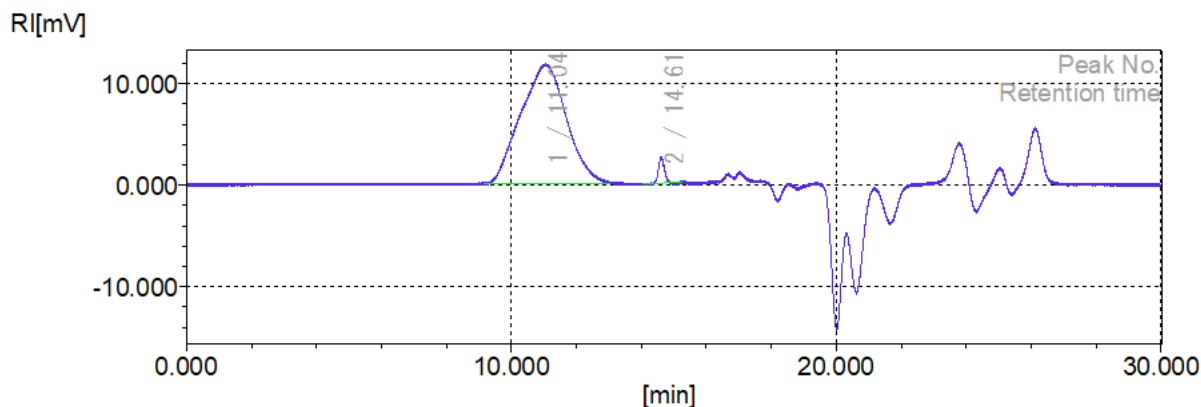


Fig S13. Raw GPC chromatogram of Table S3, Entry 1.

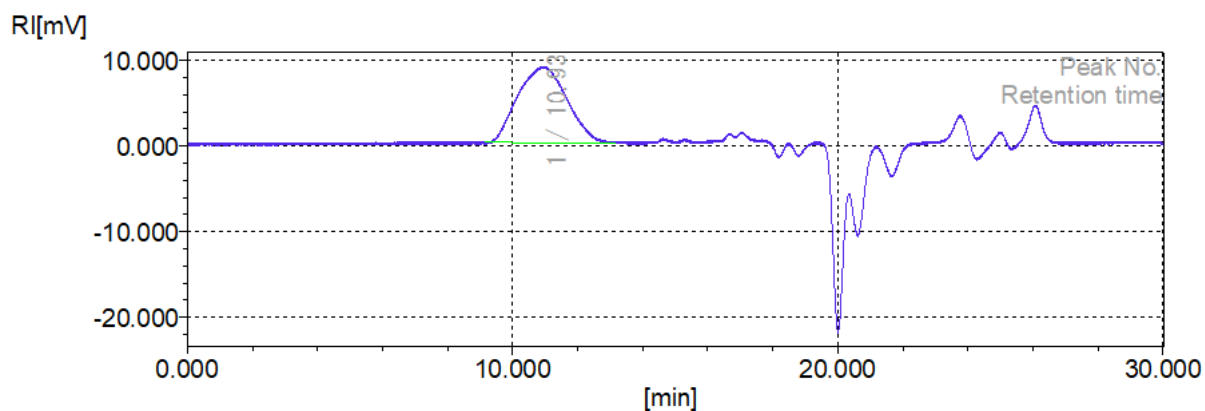


Result of molecular weight calculation (RI)

Peak 1 Base Peak

	[min]	[mV]	[mol]	Mn	202,824
Peak start	9.160	0.193	1,808,898	Mw	322,600
Peak top	11.048	11.967	244,636	Mz	480,296
Peak end	13.068	0.144	28,776	Mz+1	648,656
				Mv	322,600
Height [mV]			11.798	Mp	244,637
Area [mV*sec]			1149.205	Mz/Mw	1.49
Area% [%]			96.922	Mw/Mn	1.59
[eta]			322599.76106	Mz+1/Mw	2.01

Fig S14. Raw GPC chromatogram of Table S3, Entry 2.

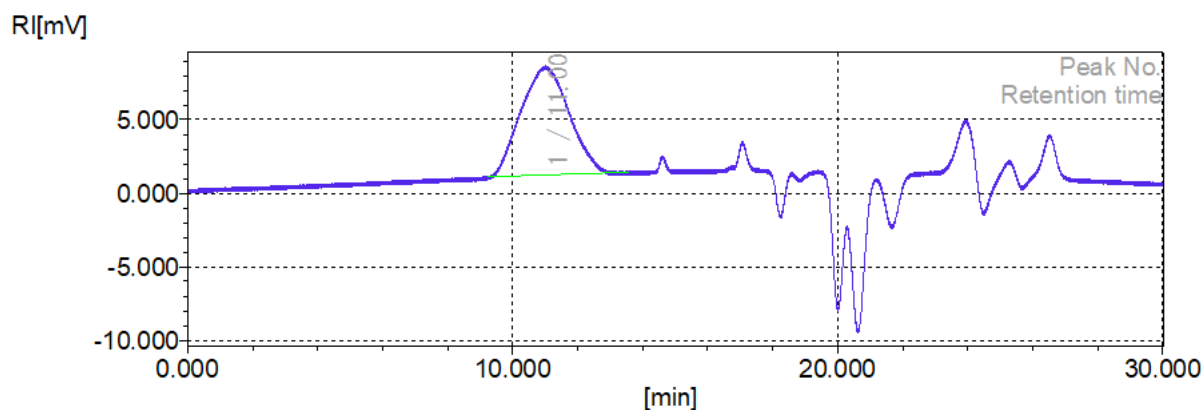


Result of molecular weight calculation (RI)

Peak 1 Base Peak

	[min]	[mV]	[mol]	Mn	213,762
Peak start	9.160	0.419	1,808,898	Mw	345,229
Peak top	10.937	9.342	275,361	Mz	510,424
Peak end	13.155	0.380	26,252	Mz+1	676,561
				Mv	345,229
Height [mV]			8.940	Mp	275,362
Area [mV*sec]			930.377	Mz/Mw	1.48
Area% [%]			100.000	Mw/Mn	1.62
[eta]			345228.61289	Mz+1/Mw	1.96

Fig S15. Raw GPC chromatogram of Table S3, Entry 2.



Result of molecular weight calculation (RI)

Peak 1 Base Peak

	[min]	[mV]	[mol]		
Peak start	9.247	1.116	1,675,703	Mn	208,230
Peak top	11.003	8.657	274,882	Mw	335,954
Peak end	13.588	1.436	19,227	Mz	494,302
				Mz+1	656,437
Height [mV]			7.412	Mv	335,954
Area [mV*sec]			763.311	Mp	274,882
Area% [%]			100.000	Mz/Mw	1.47
[eta]			335953.82354	Mw/Mn	1.61
				Mz+1/Mw	1.95

Fig S16. Raw GPC chromatogram of Table S3. Entry 4.

General Procedure D (Measurement of the Potential Window of **8**)

Linear sweep voltammetry (LSV) measurements were carried out to determine the electrochemical potential window of **8** (0.1 M in acetonitrile) in an electrochemical cell consisted of Pt button as working electrode, a leakless Ag/AgCl as reference electrode, and Pt wire as the counter electrode. The electrodes were connected to a Biologic SP150 potentiostat for electrochemical measurements. The voltage was applied from -2.5 to 2.5 V vs Ag/AgCl at 10 mV/s.

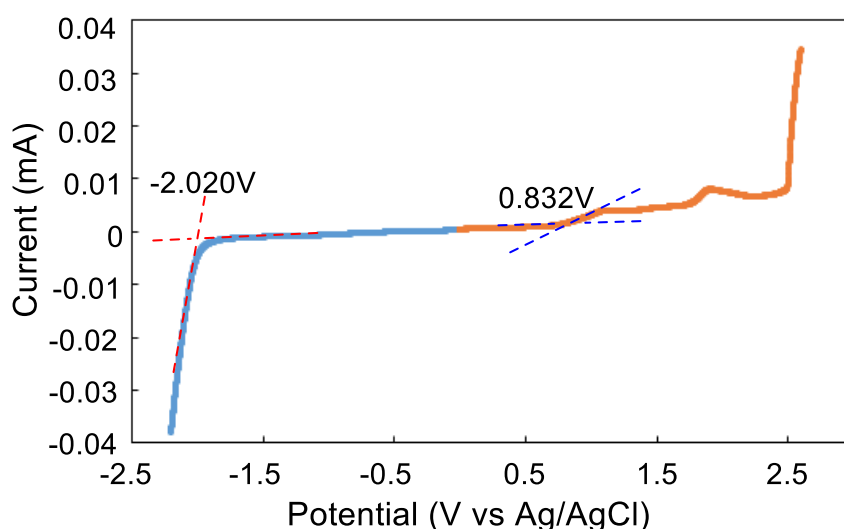


Fig S17. Linear Sweep Voltammetry of **8** in acetonitrile (0.1 M).

General Procedure E (Impedance Spectroscopy Thin Film Measurements of **8**)

Two ITO coated glass were sonicated in soap water for 15 min followed by DI water rinse and sequential sonication in acetone and isopropyl alcohol for 15 min. After drying with nitrogen flow, films substrates were treated with UV-ozone for another 20 min. Polyimide tape (Kapton tape) was used to pattern a square shape with side lengths of 5 mm to be used as the active area surface. The films were submerged into concentrated HCl for 10 min to etch ITO that are not covered with Kapton tape from the glass. Films were rinsed multiple times with DI water and after removing the Kapton tape, the substrates were cleaned one more time with the procedure explained above to remove any residual adhesive from the Kapton tape. A solution of **8** (50 mg/mL in CH₃CN) was drop cast onto the patterned square, by adding 2 μ L of solution at a time onto the active area and waiting for the solution to dry before further addition of solution. After formation of a thick layer of polymer, the film was let dry under vacuum for 6 hours. Double sided Kapton tape with approximately 100 μ m thickness was used as a spacer, before sandwiching the polymer between to the patterned ITO coated glasses. The device was hand pressed before testing to ensure a good contact between ITO and polyelectrolyte. The film thickness was measured to be 110 μ m by subtracting the two glass substrate thickness from the total device thickness. The EIS of devices was measured by VSP Potentiostat, biologic under open circuit condition with frequency range 0.1 Hz to 1 MHz with 10 mV voltage amplitude. Figure S18A shows the Nyquist plot of polyelectrolyte **8** fitted using the modified Randles circuit (Figure S18B).⁴ Using eq. 1 (Figure S18C), the ionic conductivity was calculated to be 4.5×10^{-5} S/cm, where R_{ion} is the ionic resistance, l , is the film thickness, and A is the surface area of the polymer film.

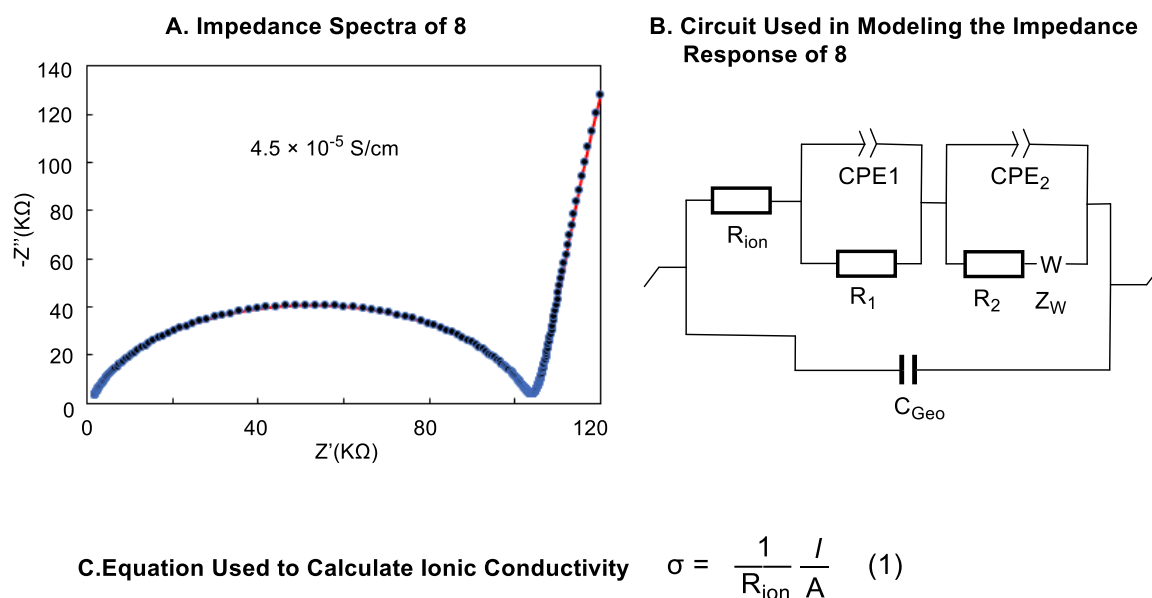


Fig S18. Ion conductivity measurement of **8** on ITO glass substrates.

General Procedure F AFM and Contact Angle Measurements of **8**

A solution of **8** (in DMF) with a concentration of 10 mg/mL was spin cast on a clean Si/SiO₂ substrate with a speed of 1000 rpm. Film morphology (Figure S19) and thickness (Figure S20) were measured using a Veeco Dimension 3100 atomic force microscope.

The hydrophobicity of **8** was measured using Ramé-hart 200 Contact Angle Goniometer. (Figure S21)

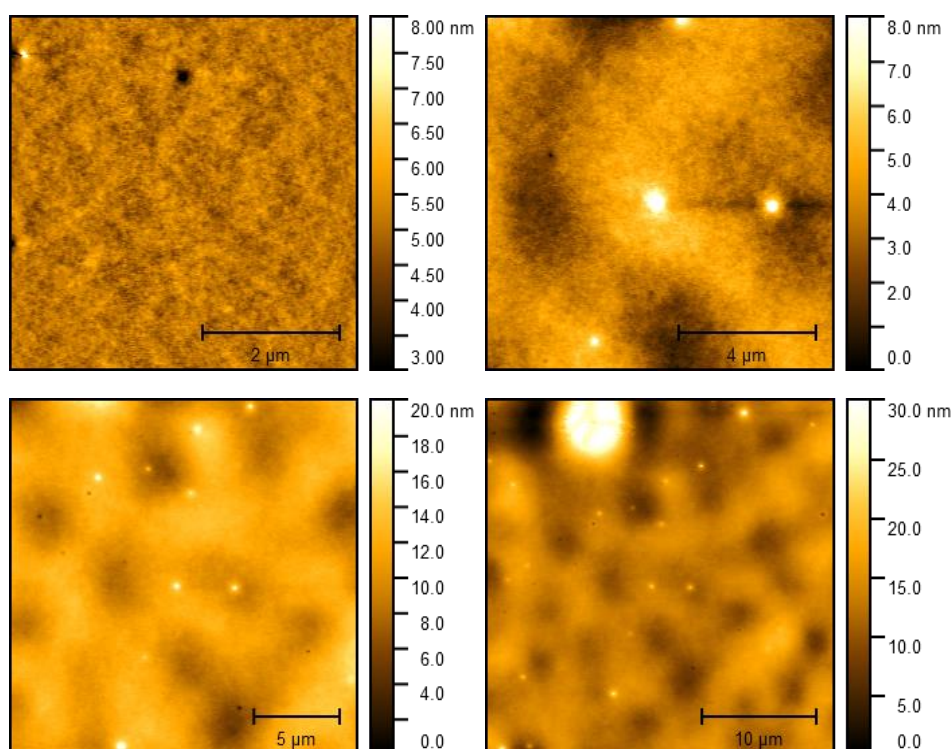


Fig S19. AFM height images of **8** surface with different scan size.

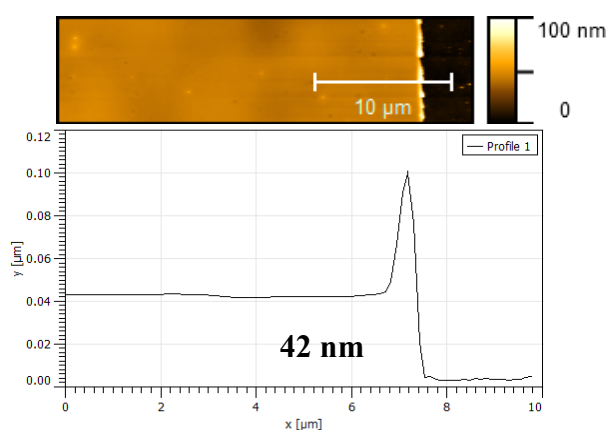


Fig S20. Film thickness of **8**. The thin film measured to an average thickness of 42 nm.

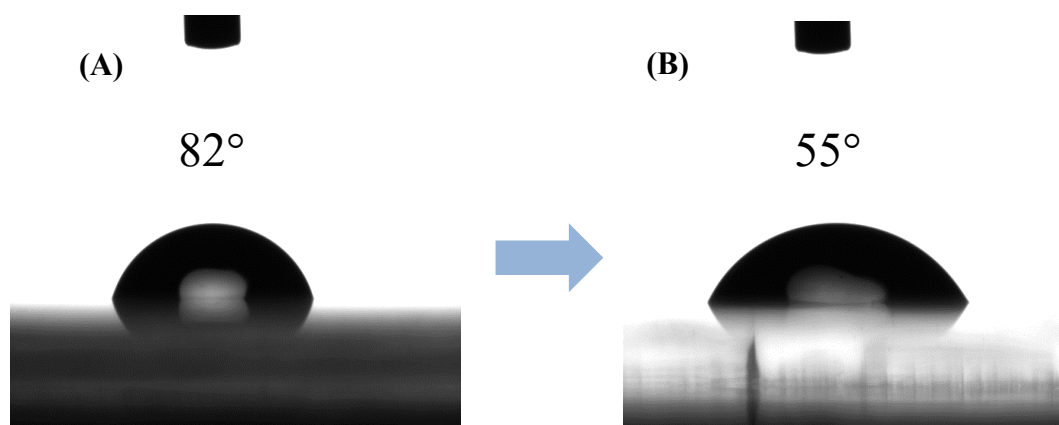
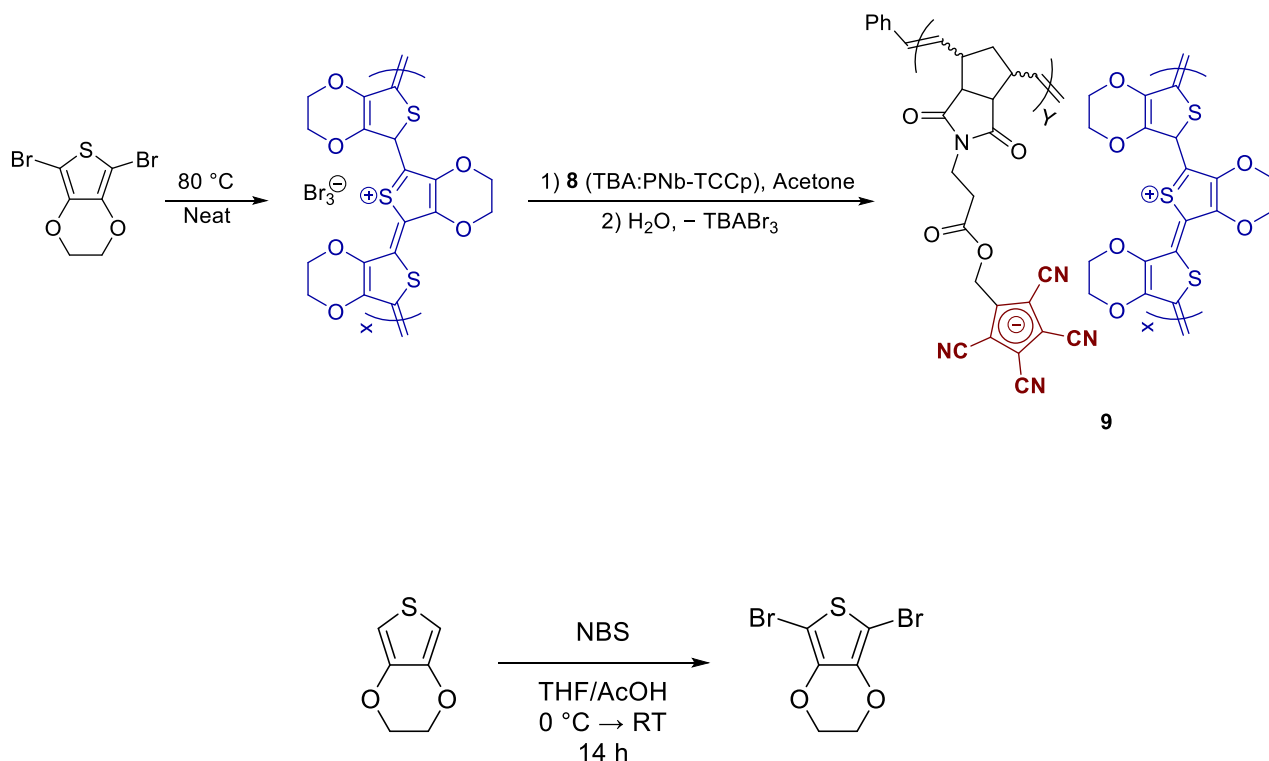
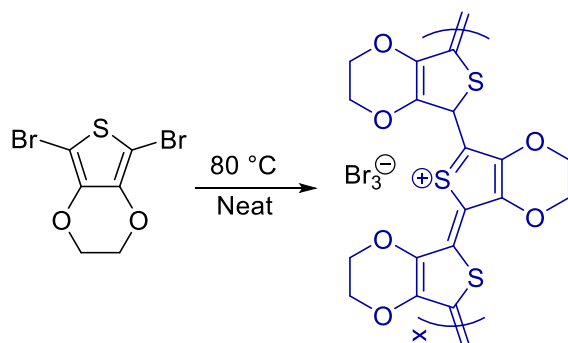


Fig S21. Water contact Angle Measurements of **8**. The water contact angle (A) was measured around 1 s after the water drop. The **8** film exhibits a hydrophilic surface with a contact angle of 82°. After several seconds, the contact angle was retaken and the surface showed dramatic decrease in contact angle, with the number of 55°.

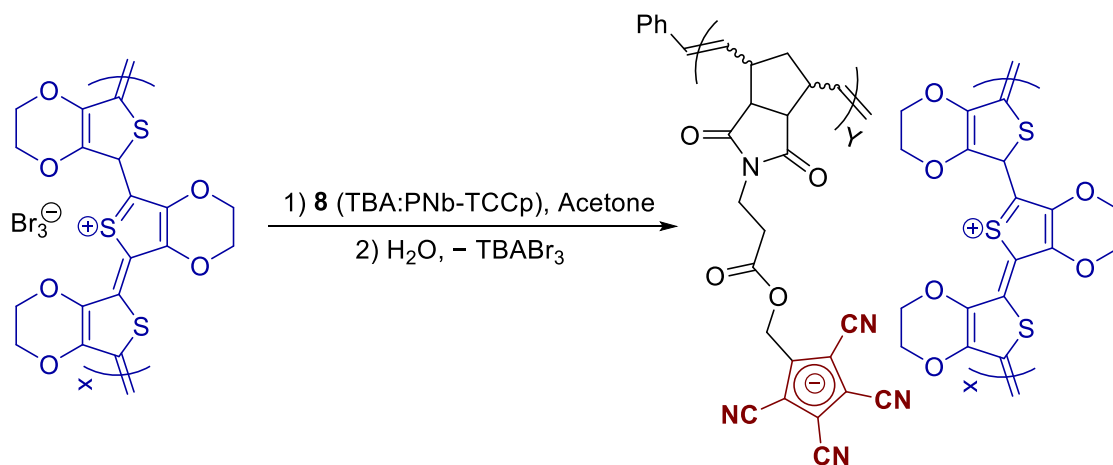
General Procedure G (Formation of PEDOT:PNb-TCCp Polymer Complex)



2,5-Dibromo-3,4-ethylenedioxythiophene (DBEDOT) was prepared following literature procedure.⁵ *N*-Bromosuccinimide (5.3 g, 2.1 eq.) was dissolved into a mixture of 1:1 (v:v) of THF:AcOH (60 mL:60mL). The reaction was bubbled with N_2 for 10 min and cooled to 0 °C. Under a stream of N_2 , 3,4-ethylenedioxythiophene (EDOT, 1.5 mL, 1.0 eq) was added drop wisely. The reaction was covered with aluminum foil and then warmed to RT to stir for 14 h. Water (230 mL) was added to the reaction and a silver/orange crystalline solid precipitated. The solids were collected by filtration and washed thoroughly with water (3 x 200 mL). The solids were collected and dried. The solids were further purified by column chromatography (2:1 Hexanes:DCM) to yield a white solid (4.0 g, 96%).



Poly(3,4-ethylenedioxythiophene) (PEDOT) was prepared following literature procedure.⁶ DBEDOT (200 mg) was heated inside a 20 mL scintillation vial at 80 °C for 4 h. The white DBEDOT powder turned to a dark blue/black powder. The resulting polymer was used without further purification.



PEDOT:P(Nb-TCCp) 9 was synthesized by first suspending PEDOT (100 mg, 1.0 eq) in acetone (1 mL) and sonicating to evenly suspend. To this suspension, a solution of **8** (100 mg, 2.0 eq) was added and sonicated for 10 min to mix. The black suspension was then precipitated into 200 mL of water and sonicated for 20 min. The resulting solids were collected by filtration and washed with water, methanol, and acetone. The solids were thoroughly dried with high vacuum for several hours to yield a dark, black powder (84.3 mg, 87%).

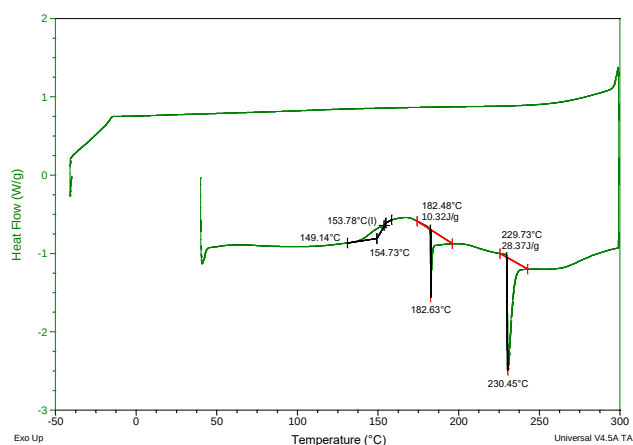


Fig S22. DSC thermogram of **PEDOT:P(Nb-TCCp) 9**, 1st heat, exo up.

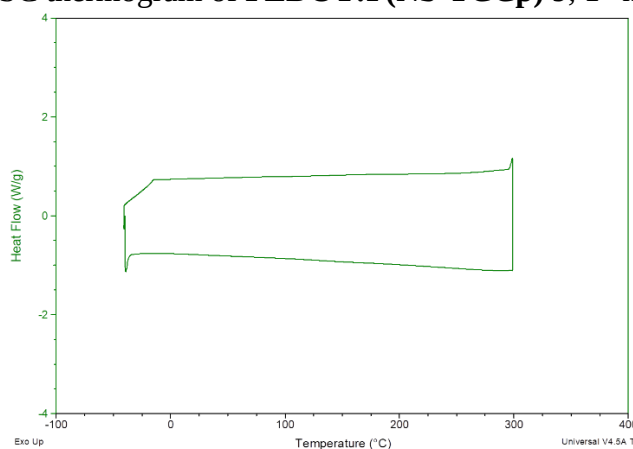


Fig S23. DSC thermogram of **PEDOT:P(Nb-TCCp) 9**, 2nd heat, exo up.

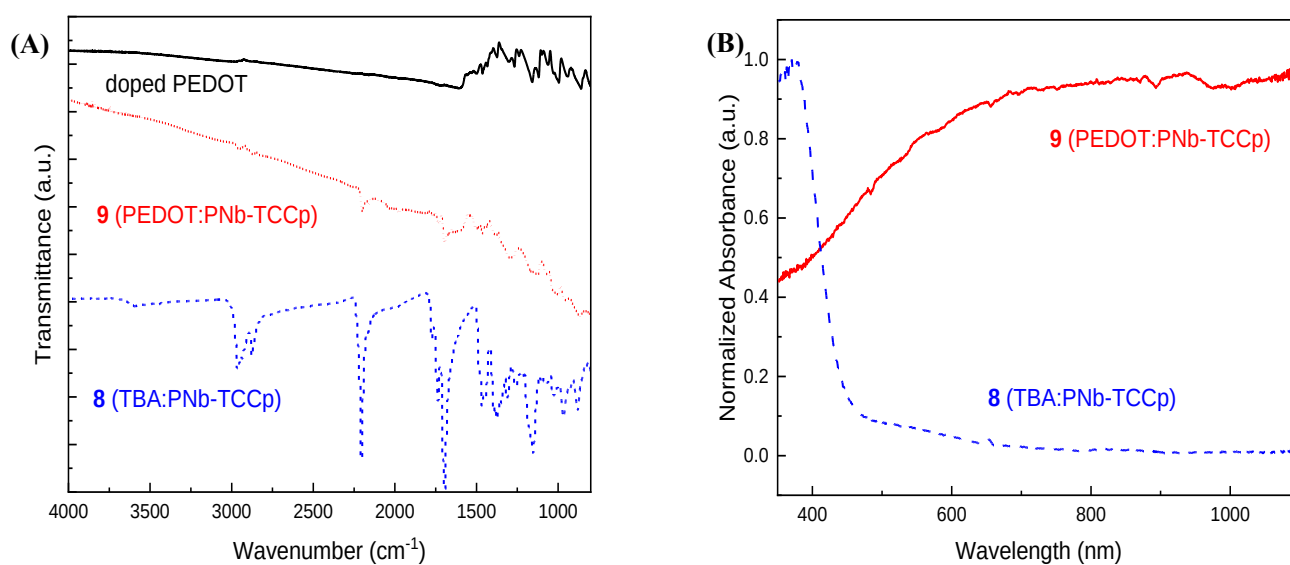


Fig S24. **A)** Non-normalized FT-IR of doped PEDOT, polymer **8**, and poly-ion complex **9**; **B)** UV/Vis of polymer **8** and poly-ion complex **9**.

General Procedure H (Electrical Conductivity Measurement of Poly-Ion Complex 9 and Doped PEDOT)

Pellets preparation: The Poly-Ion complex 9 and PEDOT powder were pressed into cylinder pellets with thickness ranging from 400 to 500 μm , and a diameter of 7.3 mm.

Electrical Conductivity Measurement: The conductivity of the pellet is measured using the van der Pauw method. Contacts were placed by touching the probe tip at four corners of the pellet with the assistance of conductive carbon paste (**Figure S25**). Two horizontal and vertical resistance were measured $R_{\perp} = (R_{AB} + R_{CD})/2$ and $R_{\parallel} = (R_{AD} + R_{BC})/2$. Specifically, R_{AB} is the resistance caused by the current passing from point A to point B while measuring the voltage between contacts C and D: $R_{AB} = V_{CD}/I_{AB}$. R_{BA} , R_{AC} , and R_{CA} were measured in the same manner. The sheet resistance R_s is then estimated by solving the equation $e^{(-\frac{\pi R_{\perp}}{R_s})} + e^{(-\frac{\pi R_{\parallel}}{R_s})} = 1$. Finally, the conductivity is calculated by $\sigma = 1/(R_s \cdot t)$ where t is the pellet thickness.

The electrical measurement was carried out using the Agilent Keysight B2902A in ambient condition. The temperature is controlled by the HFS600E-PB4 Linkam heating stage. The heating rate was kept at 20°C/min, and the pellets were allowed to reach thermal equilibrium for 5 minutes at each temperature before measurement.

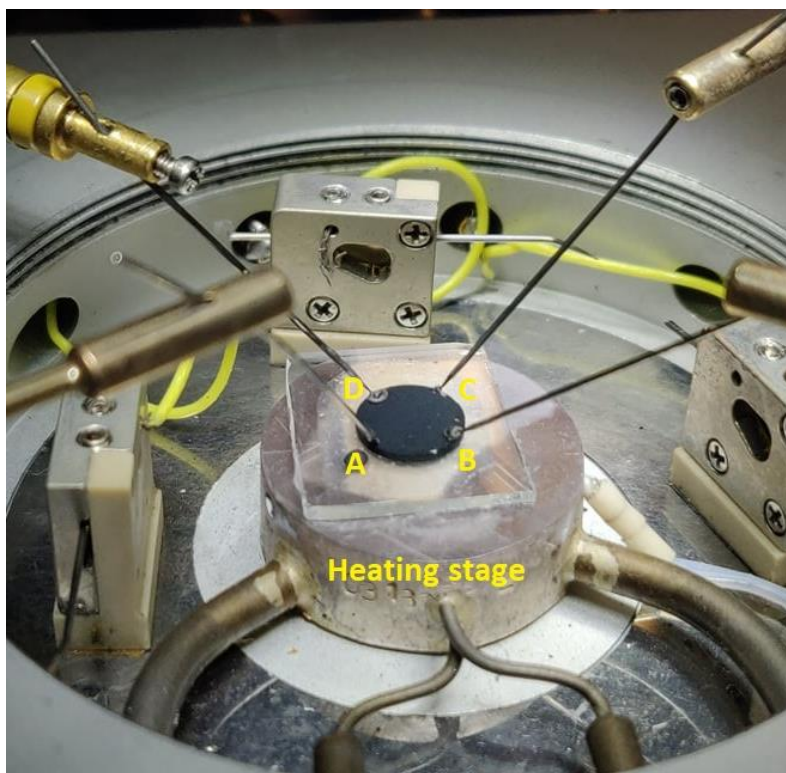


Fig S25. Setup of the in-situ temperature dependent conductivity measurement

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