

Supplemental Information

for

“Multi-ionic lithium salts increase lithium ion transference numbers in ionic liquid gel separators”

Parameswara Rao Chinnam¹, Vijay Chatare¹, Sumanth Cherreddy¹, Ramya Mantravadi¹, Michael Gau¹,
Joe Schwab² and Stephanie L. Wunder*

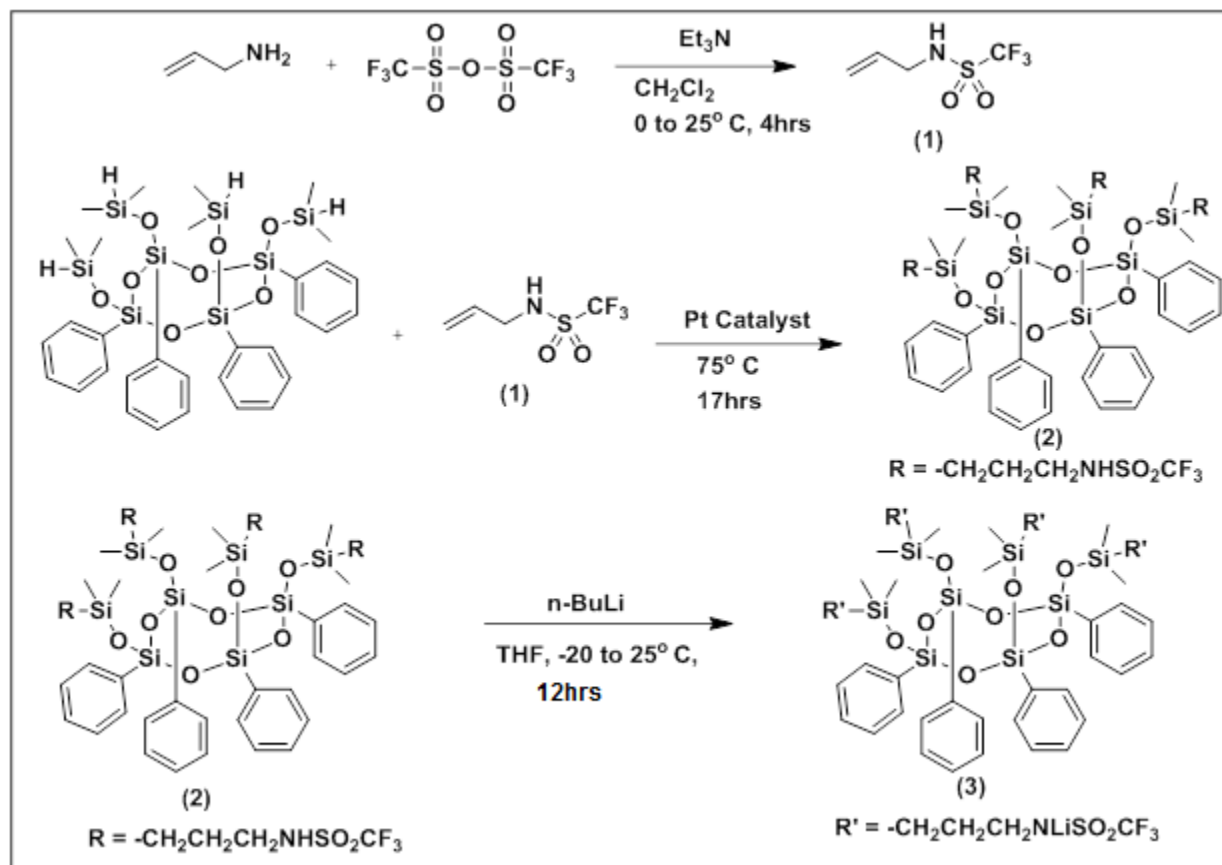
¹Department of Chemistry, Temple University, Philadelphia, PA 19122

²Hybrid Plastics, Hattiesburg, MS 39401

Table of Contents

Synthesis	2
FTIR	4
Figure S1	
Powder X-ray Diffraction.....	5
Figure S2, S3	
Thermal Gravimetric Analysis.....	7
Figure S4, S5	
Differential Scanning Calorimetry.....	9
Figures S6, S7	
Powder X-ray of blends.....	11
Figure S8	
Conductivity and t_{Li^+}	12
Figure S9 and Table S1	
Single Crystal Structure Report for 4mer-(NHTFSI) ₄	13

Synthesis of 4mer-(LiTFSD)₄ = PhSi₄O₄[(Si(CH₃)₂-R)]₄ with R = -CH₂CH₂CH₂NLiSO₂CF₃, presented in Scheme I.



Synthesis of allyltrifluoromethylsulfonamide (1)

Triflicanhydride (18 mmol) was added dropwise under an argon atmosphere to the solution of allylamine (1.0 g, 17.5 mmol) and trimethylamine (2.0 g, 20 mmol) in dry CH_2Cl_2 (40 ml) at -40 °C. To this 5.0 g of triflic anhydride (18 mmol) was added dropwise under a nitrogen atmosphere. The resultant solution was stirred for 4 h at room temperature, and volatiles were removed under reduced pressure. The viscous portion was dissolved in 30 mL of 4 M NaOH and washed with CH_2Cl_2 (3 × 25 mL). The aqueous portion was collected and then neutralized with HCl. The mixture was extracted with CH_2Cl_2 (3 × 30 mL). The organic extracts were then dried over Na_2SO_4 and filtered. The liquid product was isolated by the removal of CH_2Cl_2 under vacuum. The product was confirmed with ¹H NMR (**1**) (400 MHz, CD_3CN) δ 6.75 (s, 1H), 6.01 – 5.79 (m, 1H), 5.28 (dddd, J = 29.5, 10.3, 2.8, 1.5 Hz, 2H) and ¹⁹F NMR (376 MHz, CD_3CN) δ -77.56 (s). The NH vibration at 3315 cm^{-1} is observed in **product 1** (Figure S1).

Hydrosilylation of allyltrifluoromethylsulfonamide with 4mer-(SiH)₄ to form (2)

To a solution of 1.11 grams (2.23 mmol) of 4mer-(SiH)₄ in 12 ml of toluene, 30 μl of Pt(0) catalyst was added at 50 °C followed by the solution of allyltrifluoromethylsulfonamide(1) (1.7 grams, 8.94mmol) of in 5 ml toluene, added drop wise. The resulting solution was stirred at 75 °C for 17-19 hours. The reaction

was monitored by TLC (40% ethyl acetate in hexane) for the disappearance of reactants, after which toluene was evaporated under vacuum. The viscous crude was purified by column chromatography and the product was collected at 30 to 35% of ethyl acetate in hexane. The solvent was evaporated under reduced pressure and codistilled with CHCl_3 (3times) to remove traces of ethyl acetate. The **product 2**, T4-POSS-($\text{C}_3\text{H}_6\text{NHSO}_2\text{CF}_3$)₈, was confirmed with ^1H NMR (400 MHz, CD_3CN) δ 7.21 – 7.04 (m, 1H), 7.00 – 6.82 (m, 1H), 2.92 (t, J = 7.2 Hz, 1H), 1.73 (dt, J = 5.0, 2.5 Hz, 1H), 1.44 – 1.25 (m, 1H), 0.39 (d, J = 17.3 Hz, 1H). ^{19}F NMR (376 MHz, CD_3CN) δ -77.35 (s) and FT-IR(3315 cm^{-1}) data. The FTIR data (**Figure S1**) show the disappearance of the SiH, the appearance of the NH and appearance of the CH_2 groups during the hydrosilylation reaction.

Conversion to lithium salt, 4mer-(LiTFSI)₄ = PhSi₄O₄[(Si(CH₃)₂-R)]₄ with R = -CH₂CH₂CH₂NLiSO₂CF₃ (3)

1.4 grams of (2) was dissolved in 40 ml THF at -20 °C. To this solution 4.5 equivalents (1.3 ml) of n-butylLi were added drop-wise. The solution was stirred for 12 hours at RT. The volatiles were removed in high vacuum and the white precipitate solid was washed with excess pentane (3 X) to remove any remaining n-butyl Li. The white precipitate was filtered off, washed with THF to remove any contaminates and dried in vacuum at 70 °C for 16-18 hours to remove residual solvent (THF). **Product (3)** was confirmed with ^1H NMR and FTIR (**Figure S1**).

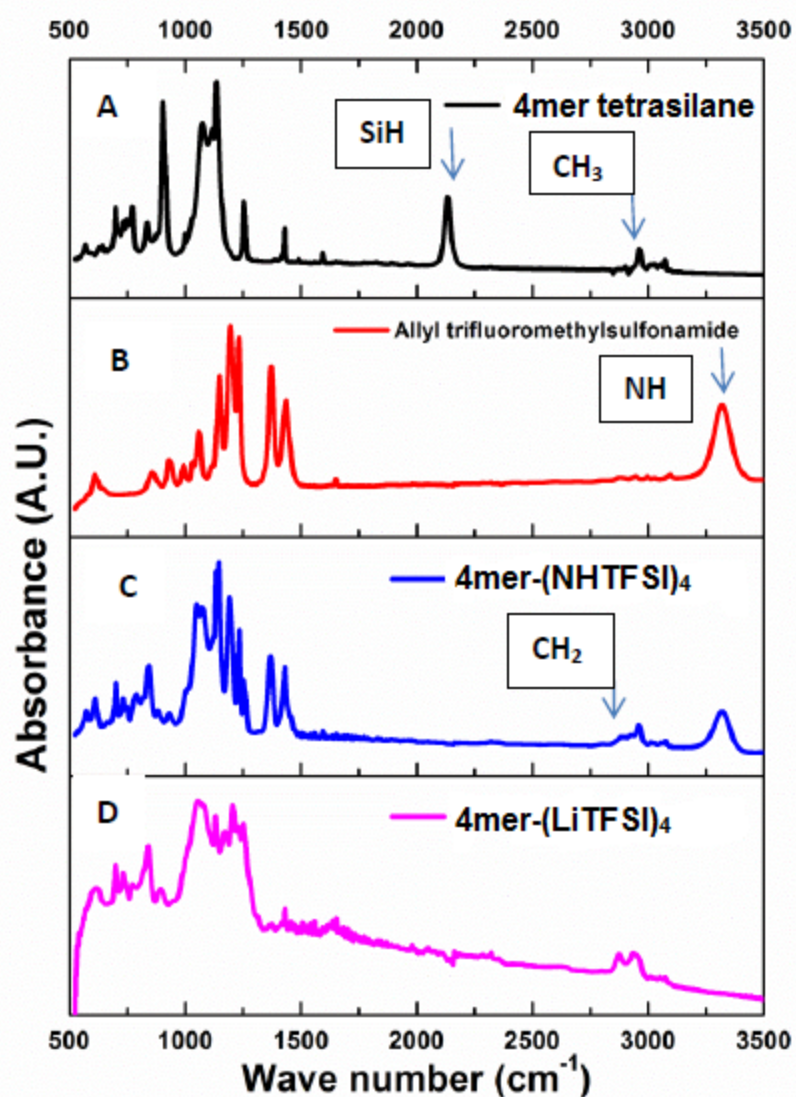


Figure S1. FTIR spectra showing: (A) Si-H and CH₃ groups of 4mer tetrasilane = 4mer-(SiH)₄; (B) NH group of allyl trifluoromethyl sulfonamide; (C) successful hydrosilylation reaction with disappearance of SiH group and formation of 4mer-(NHTFSI)₄; and (D) disappearance of NH groups due to replacement by Li and formation of 4mer-(LiTFSI)₄.

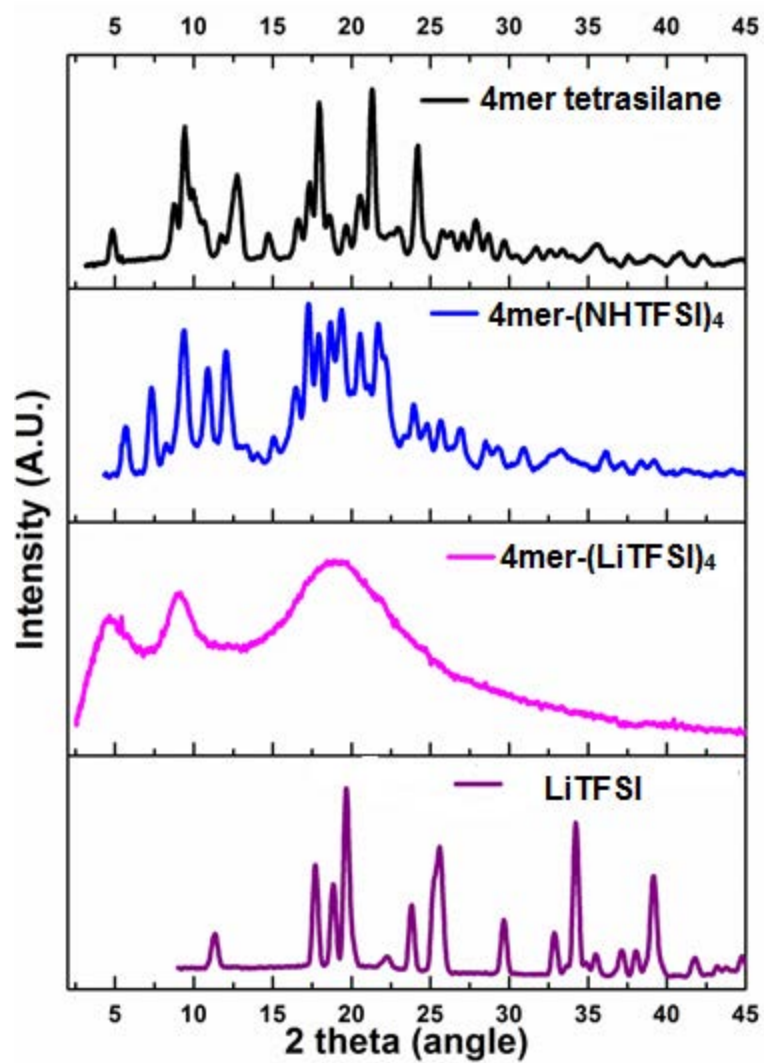


Figure S2. X-ray diffraction data for: (—) starting material, 4mer tetrasilane = 4mer-(SiH)₄; (—) amide, 4mer-(NHTFSI)₄; (—) Li salt, 4mer-(LiTFSI)₄; (—) LiTFSI, lithium bis(trifluoromethylsulfonyl)imide.

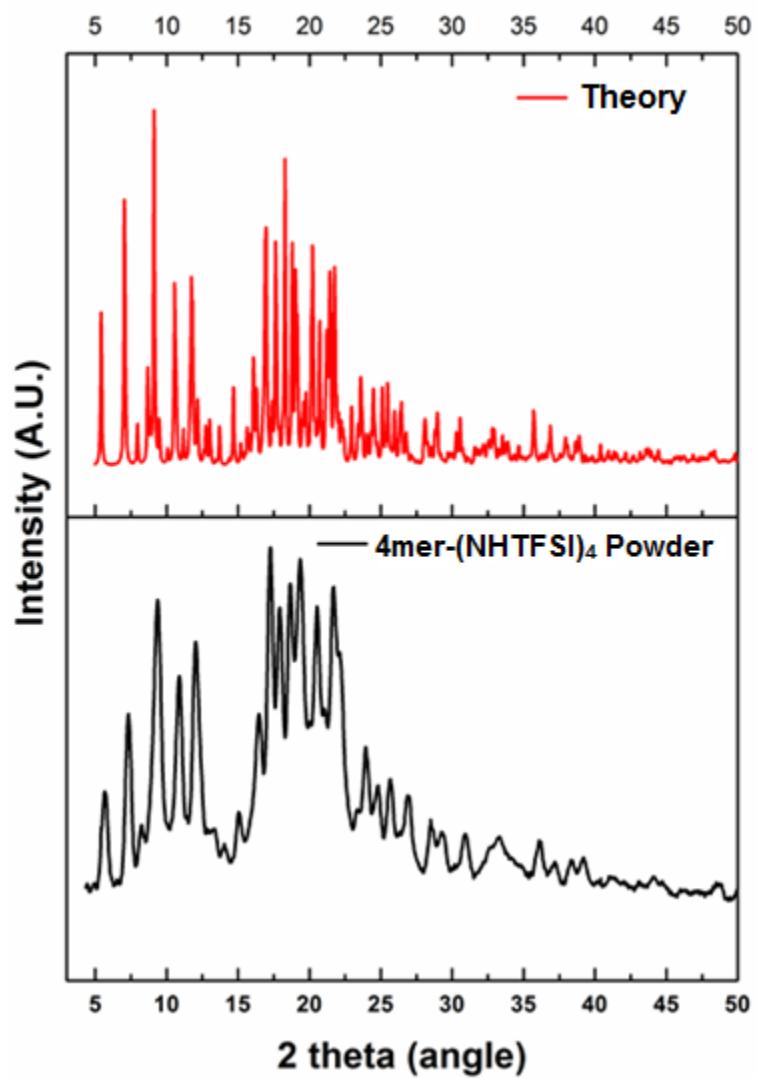


Figure S3. X-ray diffraction data for 4mer-(NHTFSI)₄ (**top**) generated from single crystal data; and (**bottom**) measured powdered pattern

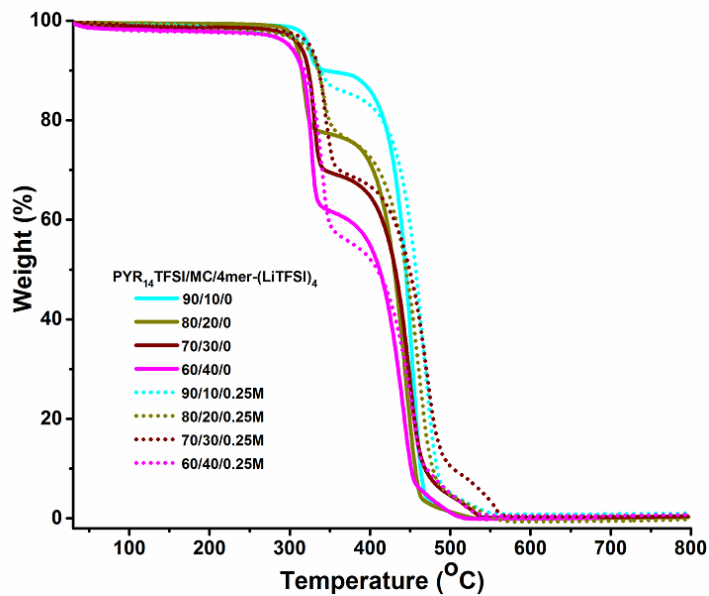
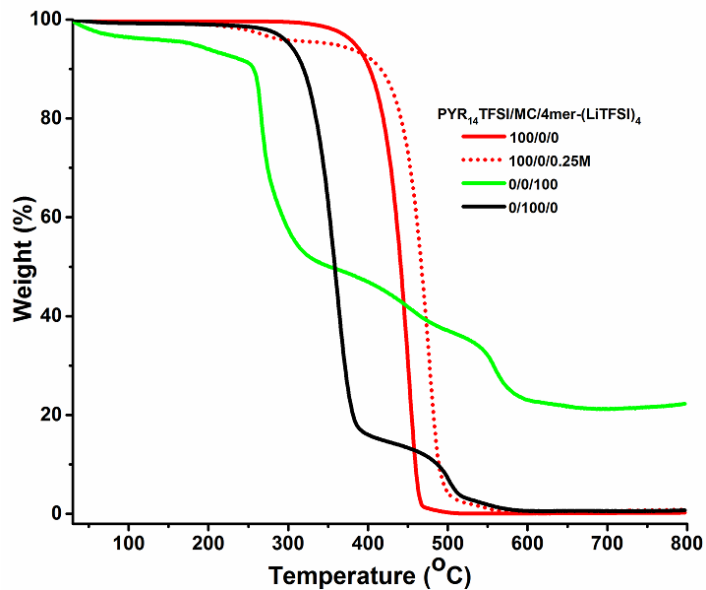


Figure S4. TGA data of neat MC, $\text{PYR}_{14}\text{TFSI}$, $4\text{mer}-(\text{LiTFSI})_4$ and $\text{PYR}_{14}\text{TFSI}/0.25\text{M } 4\text{merLiTFSI}$. Degradation peaks (T_d^{m}) were: (i) $T_d^{\text{max}} = 266\text{ }^\circ\text{C}$ (major), $450\text{ }^\circ\text{C}$ (weak) and $559\text{ }^\circ\text{C}$ (minor) for pure $4\text{mer}-(\text{LiTFSI})_4$ salt (—); (ii) $T_d^{\text{max}} = 353\text{ }^\circ\text{C}$ (major) and $500\text{ }^\circ\text{C}$ (minor) for pure MC (—); (iii) $T_d^{\text{max}} = 452\text{ }^\circ\text{C}$ (major) for pure $\text{PYR}_{14}\text{TFSI}$ (—).

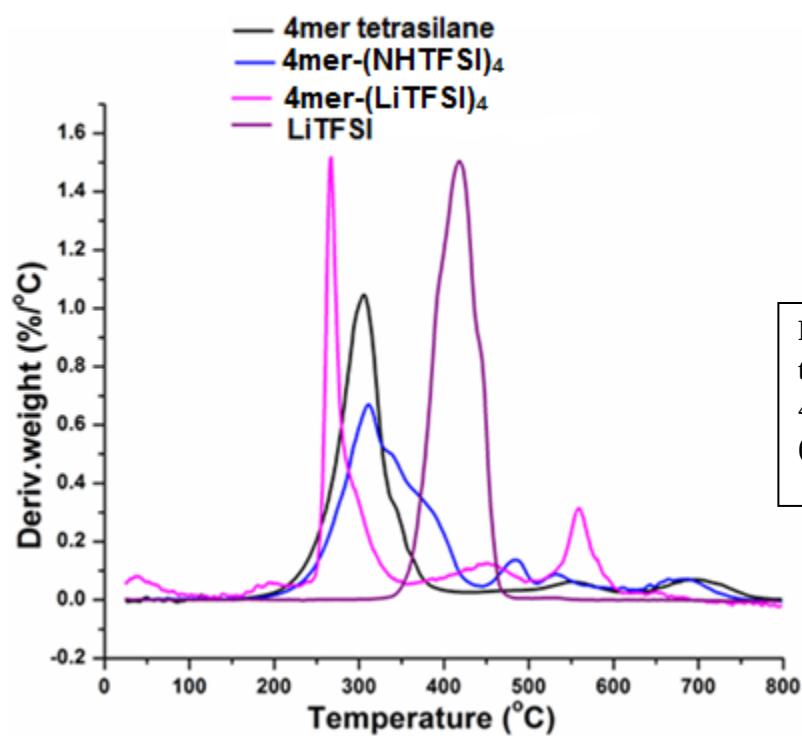
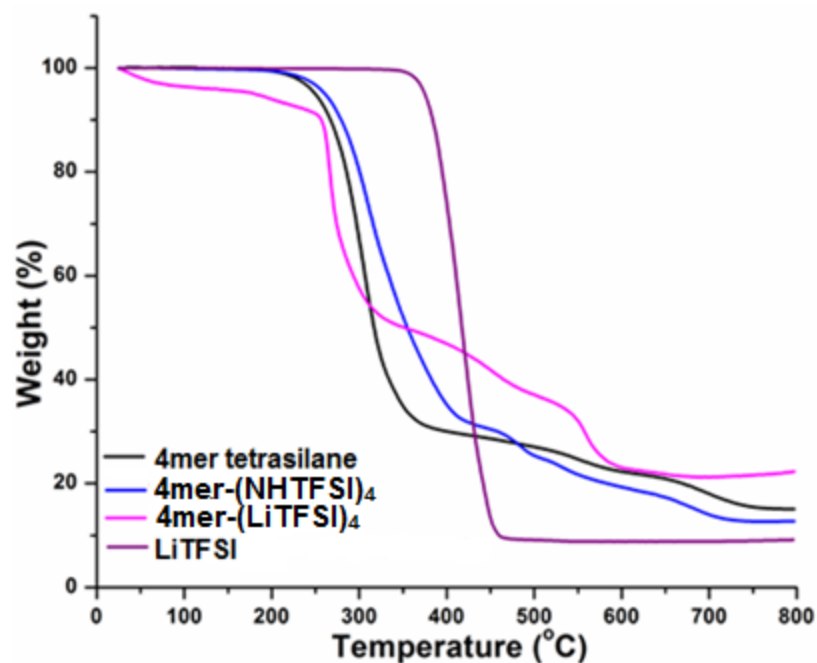


Figure S5. TGA and DTGA thermograms of LiTFSI, 4mer-(SiH)₄, 4mer-(C₃ H₆NHSO₂CF₃)₄ and 4mer-(C₃ H₆NLiSO₂CF₃)₄

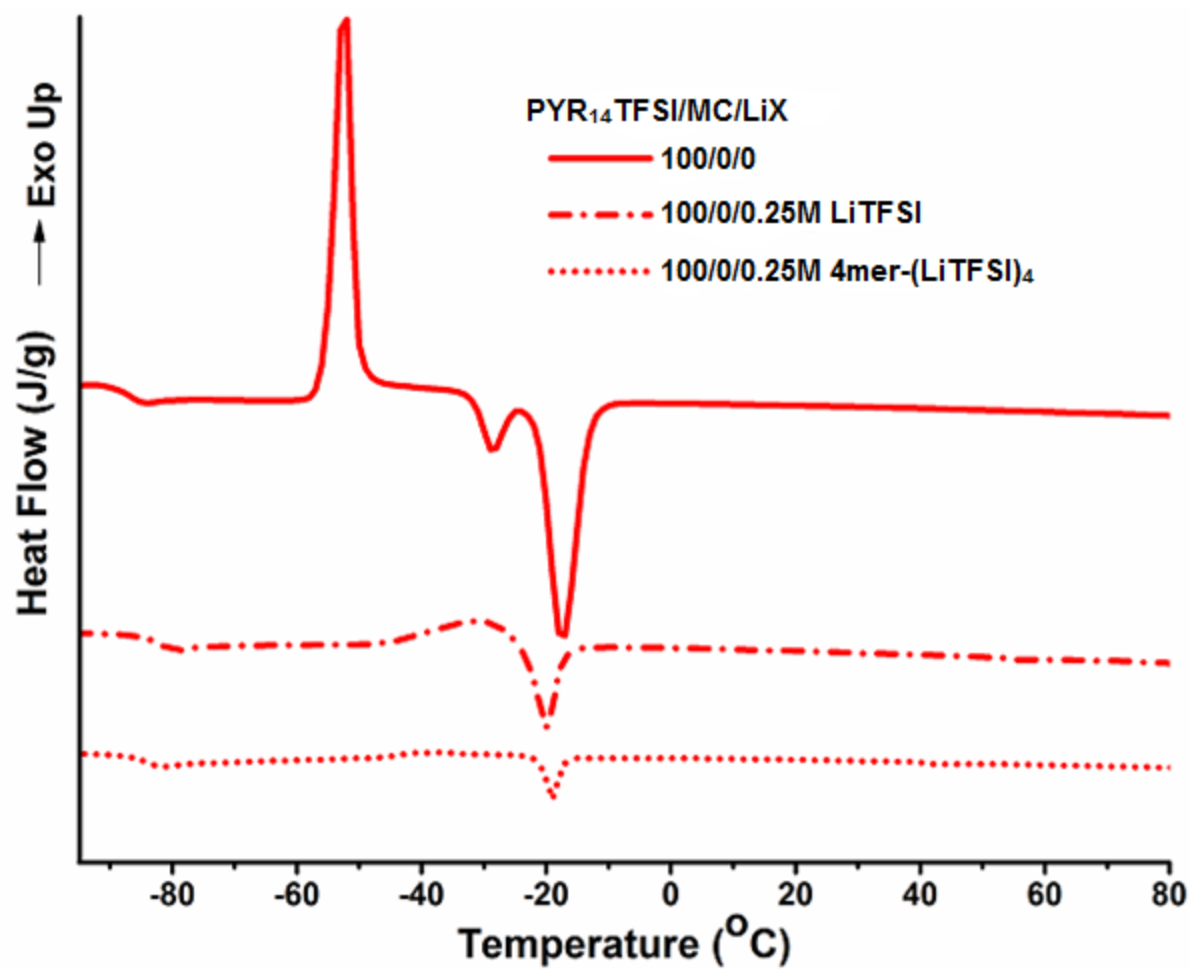


Figure S6. DSC traces (second heating cycle) of mixtures of PYR₁₄TFSI /4mer-(LiTFSI)₄ and PYR₁₄TFSI /LiTFSI, 0.25M

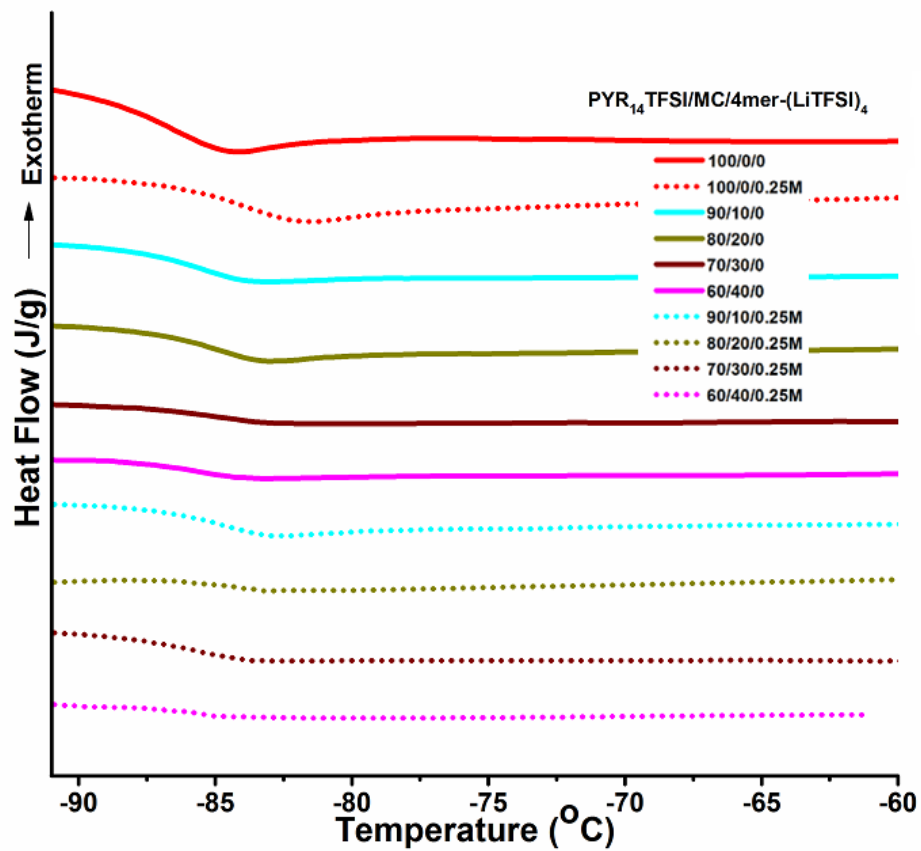


Figure S7. DSC traces of PYR₁₄TFSI/MC/0.25M 4mer-(LiTFSI)₄ ion-gels from -60 to -90 °C

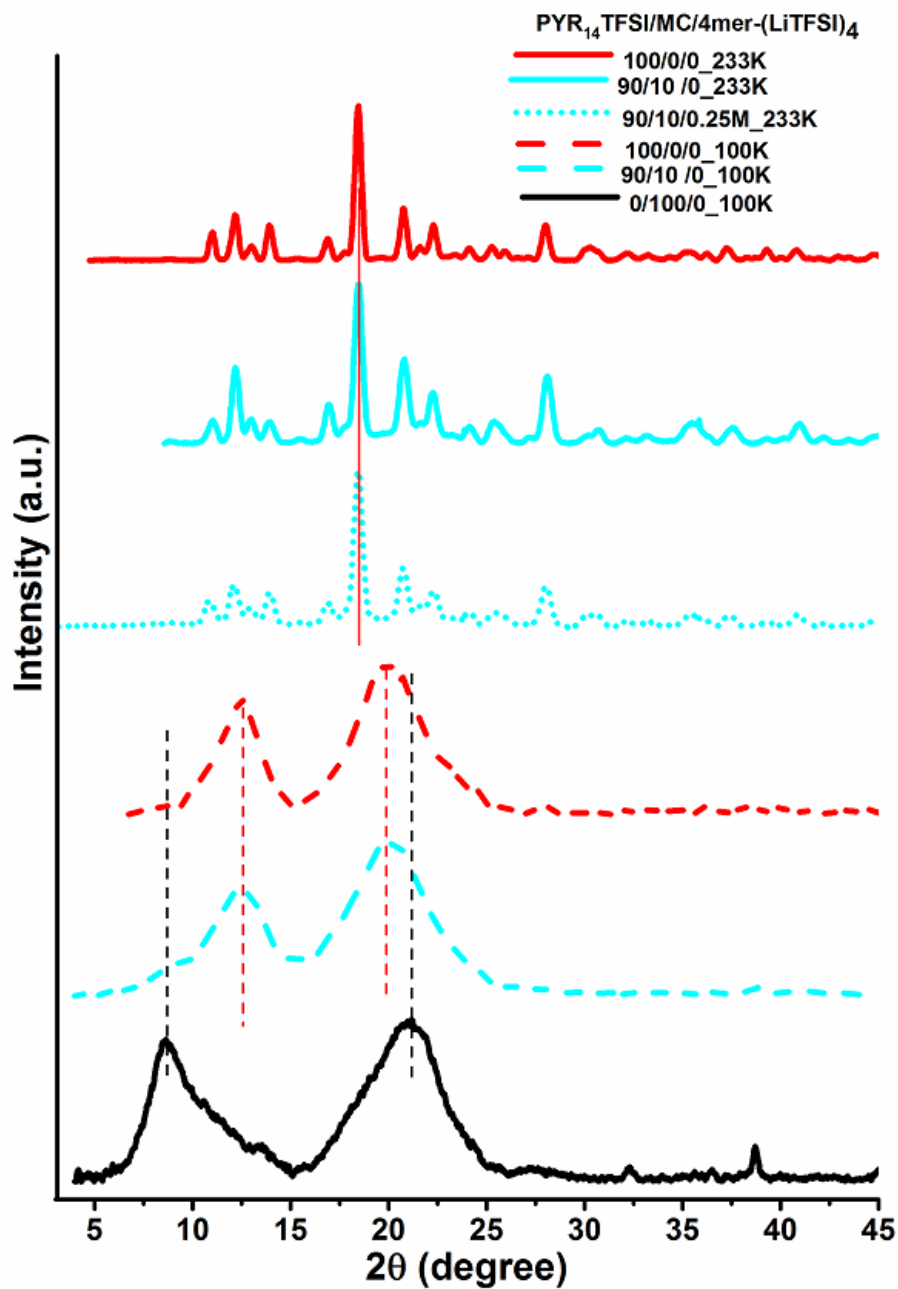


Figure S8. X-ray diffraction data showing samples that have been cooled to -100°C , where supercooling keeps the $\text{PYR}_{14}\text{TFSI}$ amorphous and those reheated to -40°C , where the $\text{PYR}_{14}\text{TFSI}$ crystallizes but does not remelt. In the 90/10 $\text{PYR}_{14}\text{TFSI}/\text{MC}$ or 90/10 $\text{PYR}_{14}\text{TFSI}/\text{MC}/0.25\text{M } 4\text{mer}-(\text{LiTFSI})_4$, the MC peaks are too weak to be seen, but the $\text{PYR}_{14}/\text{TFSI}$ and $\text{PYR}_{14}/\text{TFSI}/0.25\text{M } 4\text{mer}-(\text{LiTFSI})_4$ are distinct, crystalline phase-separated phases.

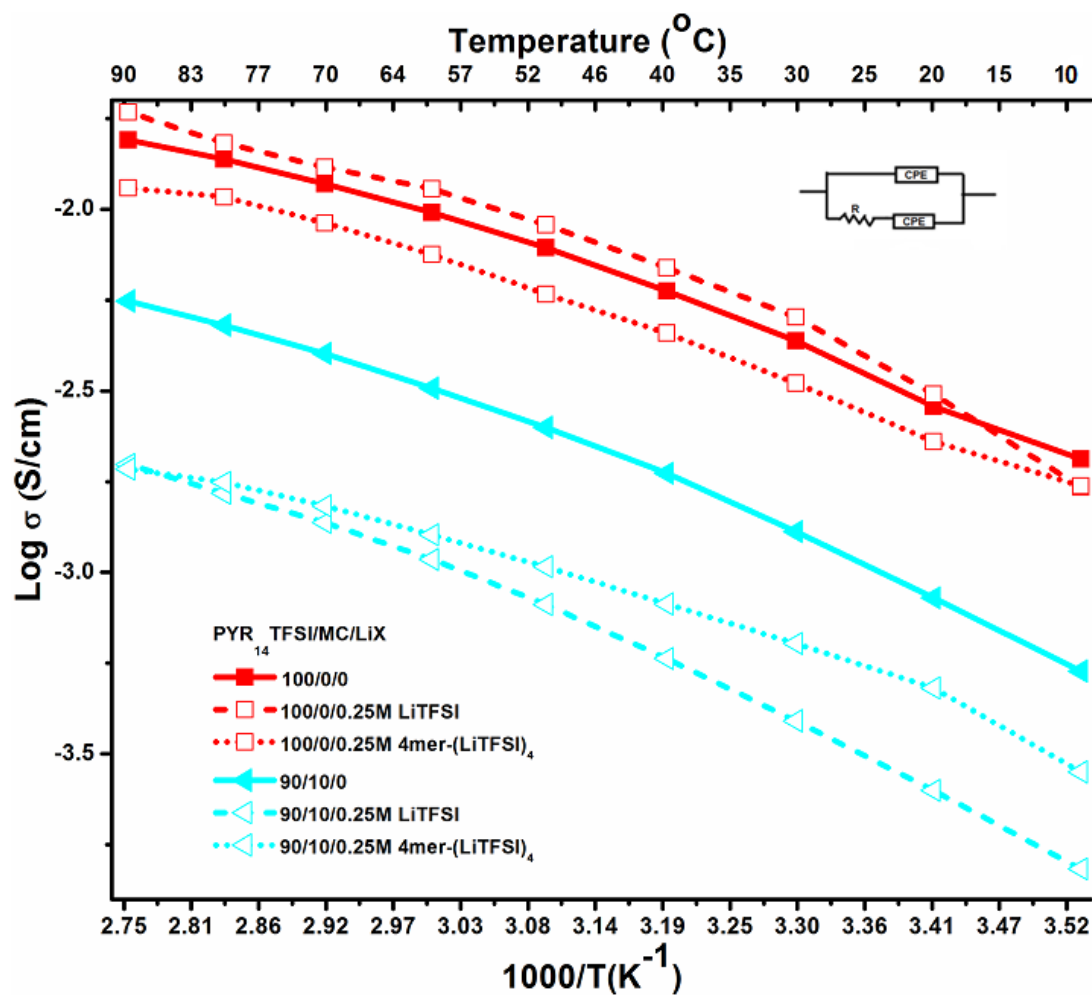


Figure S9. Comparison of conductivities for $\text{PYR}_{14}\text{TFSI/MC/4mer-(LiTFSI)}_4$ and $\text{PYR}_{14}\text{TFSI/MC/LiTFSI}$

Table S1. Comparison of t_{Li^+} for $\text{PYR}_{14}\text{TFSI/MC/4mer-(LiTFSI)}_4$ and $\text{PYR}_{14}\text{TFSI/MC/LiTFSI}$		
	LiX	
	4merLiTFSI	LiTFSI
$\text{PYR}_{14}\text{TFSI/MC/LiX}$		
100/0/0.25M	0.08, 0.10	0.10
90/10/0.25M	0.19	0.16
80/20/0.25M	0.35	0.18
70/30/0.25M	0.36	0.21
60/40/0.25M	0.39	0.23

Crystal Structure Report for 4mer-(NHTFSI)₄

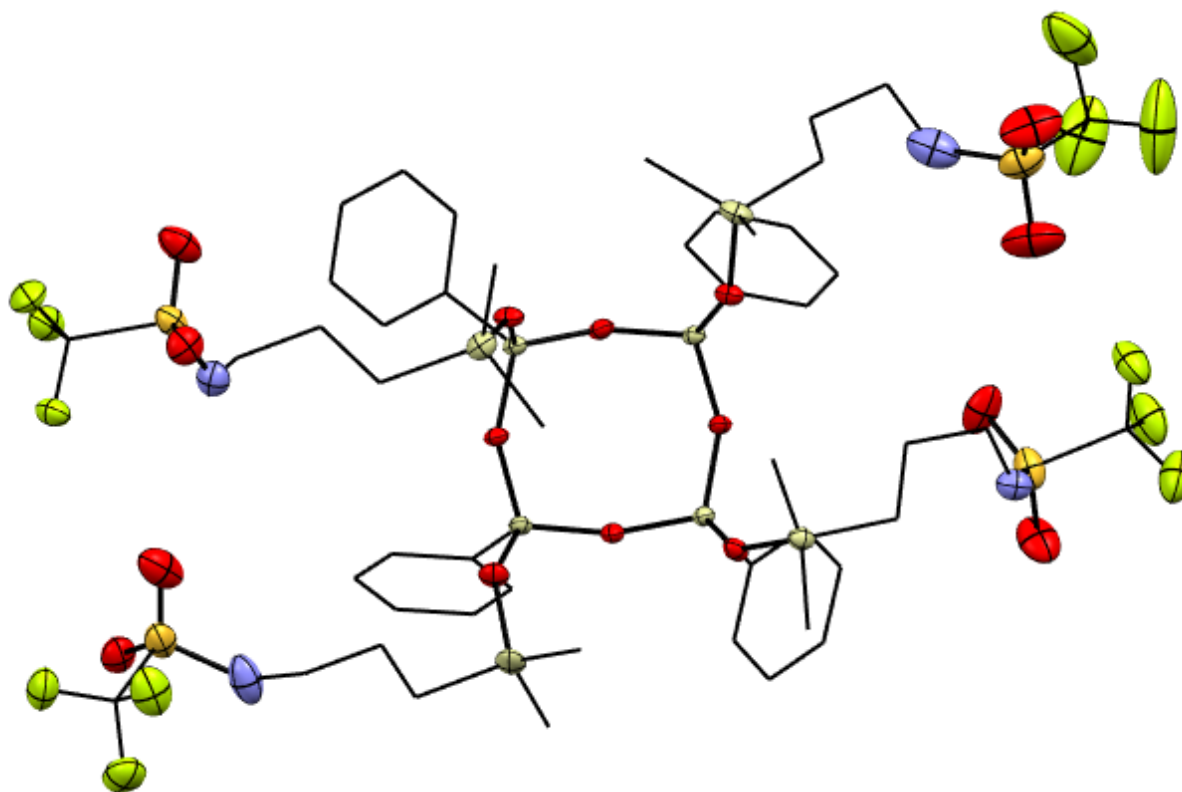


Figure S10. Crystal structure of 4mer-(NHTFSI)₄. H atoms have been omitted for clarity and C atoms are shown as wireframe. Ellipsoids are shown at 30% probability.

A specimen of C₄₈H₇₂F₁₂N₄O₁₆S₄Si₈, approximate dimensions 0.070 mm x 0.190 mm x 0.335 mm, was used for the X-ray crystallographic analysis. The X-ray intensity data were measured.

The total exposure time was 9.81 hours. The frames were integrated with the Bruker SAINT software package using a narrow-frame algorithm. The integration of the data using triclinic unit cell yielded a total of 28461 reflections to a maximum θ angle of 23.26° (0.90 Å resolution), of which 10298 were independent (average redundancy 2.764, completeness = 99.8%, R_{int} = 4.51%, R_{sig} = 5.83%) and 7086 (68.81%) were greater than $2\sigma(F^2)$. The final cell constants of a = 11.371(3) Å, b = 16.642(4) Å, c = 19.433(5) Å, α = 89.699(14)°, β = 85.466(5)°, γ = 78.305(4)°, volume = 3589.6(15) Å³, are based upon the refinement of the XYZ-centroids of 5984 reflections above $20\sigma(I)$ with $4.841^\circ < 2\theta < 47.90^\circ$. Data were corrected for absorption effects using the multi-scan method (SADABS). The ratio of minimum to maximum apparent transmission was 0.877. The calculated minimum and maximum transmission coefficients (based on crystal size) are 0.6541 and 0.7456.

The structure was solved and refined using the Bruker SHELXTL Software Package, using the space group P -1, with $Z = 2$ for the formula unit, C₄₈H₇₂F₁₂N₄O₁₆S₄Si₈. The final anisotropic full-matrix least-squares refinement on F^2 with 837 variables converged at R_1 = 7.59%, for the observed data and wR_2 = 21.85% for all data. The goodness-of-fit was 1.047. The largest peak in the final difference electron

density synthesis was $1.279 \text{ e}^-/\text{\AA}^3$ and the largest hole was $-0.656 \text{ e}^-/\text{\AA}^3$ with an RMS deviation of $0.094 \text{ e}^-/\text{\AA}^3$. On the basis of the final model, the calculated density was 1.427 g/cm^3 and $F(000)$, 1600 e^- .

Table S2. Sample and crystal data for 4merNH-TFSI.

Identification code	4mer-(NHTFSI) ₄	
Chemical formula	C ₄₈ H ₇₂ F ₁₂ N ₄ O ₁₆ S ₄ Si ₈	
Formula weight	1542.05 g/mol	
Temperature	100(2) K	
Wavelength	0.71073 Å	
Crystal size	0.070 x 0.190 x 0.335 mm	
Crystal system	triclinic	
Space group	P -1	
Unit cell dimensions	a = 11.371(3) Å	$\alpha = 89.699(14)^\circ$
	b = 16.642(4) Å	$\beta = 85.466(5)^\circ$
	c = 19.433(5) Å	$\gamma = 78.305(4)^\circ$
Volume	3589.6(15) Å ³	
Z	2	
Density (calculated)	1.427 g/cm ³	
Absorption coefficient	0.358 mm ⁻¹	
F(000)	1600	

Table S3. Data collection and structure refinement for 4mer-(NHTFSI)₄.

Theta range for data collection	1.05 to 23.26°	
Index ranges	-12 ≤ h ≤ 12, -18 ≤ k ≤ 18, -21 ≤ l ≤ 20	
Reflections collected	28461	
Independent reflections	10298 [R(int) = 0.0451]	
Coverage of independent reflections	99.8%	
Absorption correction	multi-scan	
Max. and min. transmission	0.7456 and 0.6541	
Structure solution technique	direct methods	
Structure solution program	SHELXS-97 (Sheldrick, 2008)	
Refinement method	Full-matrix least-squares on F ²	
Refinement program	SHELXL-2014/7 (Sheldrick, 2014)	
Function minimized	$\sum w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	10298 / 1 / 837	
Goodness-of-fit on F ²	1.047	
Final R indices	7086 data; I > 2σ(I)	R1 = 0.0759, wR2 = 0.1930
	all data	R1 = 0.1137, wR2 = 0.2185
Weighting scheme	w = 1/[σ ² (F _o ²) + (0.1054P) ² + 11.3886P]	

Largest diff. peak and hole
R.M.S. deviation from mean

where $P=(F_o^2+2F_c^2)/3$
 1.279 and -0.656 eÅ⁻³
 0.094 eÅ⁻³

Table S4. Atomic coordinates and equivalent isotropic atomic displacement parameters (Å²) for 4mer-(NHTFSI)₄.

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	y/b	z/c	U(eq)
Si1	0.98127(13)	0.50725(9)	0.19917(8)	0.0256(4)
Si2	0.26405(13)	0.27975(9)	0.21293(8)	0.0261(4)
Si3	0.25769(13)	0.47165(9)	0.22065(8)	0.0255(4)
Si4	0.98215(13)	0.31816(10)	0.21570(8)	0.0259(4)
Si5	0.25556(14)	0.55882(10)	0.36171(9)	0.0322(4)
S8	0.85666(17)	0.95471(12)	0.43717(10)	0.0525(5)
Si7	0.76059(16)	0.57556(12)	0.30161(10)	0.0443(5)
Si8	0.93383(16)	0.27465(11)	0.36764(9)	0.0388(5)
Si9	0.44290(17)	0.22955(12)	0.32155(11)	0.0480(5)
C11	0.9396(5)	0.5616(4)	0.1186(3)	0.0333(14)
C12	0.8789(5)	0.5285(4)	0.0700(3)	0.0411(16)
C16	0.9701(6)	0.6378(4)	0.1048(4)	0.0486(19)
C14	0.8782(6)	0.6463(6)	0.9989(4)	0.067(3)
C13	0.8464(6)	0.5720(5)	0.0109(4)	0.055(2)
C15	0.9401(6)	0.6808(5)	0.0448(4)	0.063(2)
C21	0.3372(5)	0.2263(3)	0.1333(3)	0.0275(13)
C25	0.4568(5)	0.2209(4)	0.0246(3)	0.0383(15)
C26	0.4016(5)	0.2627(4)	0.0830(3)	0.0311(14)
C23	0.3833(7)	0.1037(4)	0.0599(4)	0.059(2)
C24	0.4482(6)	0.1414(4)	0.0134(4)	0.0477(18)
C22	0.3290(6)	0.1450(4)	0.1198(4)	0.0508(19)
C31	0.3557(5)	0.5129(3)	0.1546(3)	0.0304(14)
C32	0.4768(5)	0.4768(4)	0.1444(4)	0.0432(17)
C34	0.5110(7)	0.5742(5)	0.0583(4)	0.055(2)
C33	0.5542(6)	0.5068(5)	0.0960(4)	0.0525(19)
C36	0.3151(6)	0.5788(4)	0.1142(3)	0.0434(17)
C35	0.3917(6)	0.6104(4)	0.0666(4)	0.0519(19)
C41	0.9140(5)	0.2631(4)	0.1528(3)	0.0303(14)
C42	0.7955(6)	0.2938(5)	0.1378(4)	0.058(2)
C44	0.7970(7)	0.1827(6)	0.0626(4)	0.062(2)
C46	0.9719(7)	0.1929(5)	0.1194(4)	0.0543(19)
C45	0.9140(8)	0.1512(5)	0.0735(5)	0.073(3)
C83A	0.7395(7)	0.2530(6)	0.0932(5)	0.070(2)

	x/a	y/b	z/c	U(eq)
O3	0.2819(3)	0.3735(2)	0.2105(2)	0.0319(10)
O2	0.1186(3)	0.5117(2)	0.2109(2)	0.0284(9)
O1	0.9684(3)	0.4131(2)	0.1922(2)	0.0320(9)
O4	0.1223(3)	0.2764(2)	0.2186(2)	0.0289(9)
O5	0.8994(3)	0.5502(2)	0.2654(2)	0.0334(10)
O6	0.2885(3)	0.4919(2)	0.2971(2)	0.0321(9)
O7	0.9140(3)	0.3144(2)	0.2911(2)	0.0339(10)
O8	0.3190(4)	0.2359(3)	0.2804(2)	0.0387(10)
C84	0.8840(6)	0.8448(5)	0.4202(4)	0.0475(18)
C83	0.9519(10)	0.0200(5)	0.3252(4)	0.077(3)
N8	0.9616(6)	0.9867(4)	0.3944(3)	0.0604(17)
O81	0.8741(5)	0.9587(3)	0.5082(3)	0.0661(15)
O82	0.7454(5)	0.9840(4)	0.4116(3)	0.0796(18)
F81	0.9919(3)	0.8098(2)	0.4373(2)	0.0562(11)
F83	0.8781(4)	0.8294(3)	0.3541(2)	0.0696(13)
F82	0.8039(4)	0.8112(3)	0.4572(3)	0.0746(14)
C71	0.6946(8)	0.6796(5)	0.2691(5)	0.080(3)
C72	0.6487(8)	0.6819(5)	0.1987(5)	0.079(3)
C73	0.6084(9)	0.7709(7)	0.1720(6)	0.111(4)
N7	0.7143(8)	0.8044(6)	0.1607(4)	0.101(3)
S7	0.7269(2)	0.89014(18)	0.20254(13)	0.0818(7)
O71	0.6550(6)	0.9046(6)	0.2640(3)	0.119(3)
O72	0.8509(7)	0.8828(6)	0.2017(5)	0.141(3)
C51	0.2402(6)	0.6654(4)	0.3276(4)	0.0439(17)
C53	0.1245(6)	0.7845(4)	0.2621(4)	0.0471(17)
C52	0.1368(6)	0.6956(4)	0.2828(4)	0.0445(17)
C54	0.1698(9)	0.9370(5)	0.1403(5)	0.068(2)
N5	0.2267(5)	0.8015(4)	0.2202(3)	0.0535(16)
S5	0.2260(2)	0.82616(12)	0.14143(11)	0.0626(6)
O52	0.3467(6)	0.8193(4)	0.1132(3)	0.094(2)
O51	0.1409(6)	0.7934(4)	0.1094(3)	0.085(2)
F53	0.2360(6)	0.9736(3)	0.1792(3)	0.1044(19)
F52	0.0592(5)	0.9565(3)	0.1672(3)	0.0939(17)
F51	0.1753(6)	0.9653(3)	0.0774(3)	0.1031(19)
C91	0.4854(7)	0.1206(5)	0.3474(5)	0.065(2)
C92	0.3881(7)	0.0955(5)	0.3915(5)	0.067(2)
C94	0.4459(8)	0.8032(5)	0.4382(4)	0.059(2)
C93	0.4196(9)	0.0073(6)	0.4198(5)	0.088(3)
S9	0.3746(2)	0.86882(13)	0.37055(12)	0.0653(6)
O92	0.4134(5)	0.8251(3)	0.3089(3)	0.0661(15)

	x/a	y/b	z/c	U(eq)
O91	0.2551(6)	0.8852(4)	0.3937(3)	0.094(2)
F93	0.4228(5)	0.7307(3)	0.4390(3)	0.0788(14)
F92	0.5656(5)	0.7946(3)	0.4274(3)	0.0908(17)
F91	0.4141(5)	0.8387(3)	0.4998(3)	0.0948(17)
C8A	0.3804(6)	0.5400(4)	0.4191(3)	0.0456(17)
C5A	0.4053(7)	0.3008(4)	0.3966(4)	0.060(2)
C7A	0.1146(6)	0.5459(5)	0.4104(4)	0.0512(18)
C6A	0.5651(7)	0.2565(6)	0.2621(5)	0.081(3)
C3A	0.0530(7)	0.3165(6)	0.4054(4)	0.071(3)
C4A	0.7908(7)	0.3059(7)	0.4218(4)	0.077(3)
C2A	0.7686(7)	0.5840(6)	0.3952(4)	0.067(2)
C1A	0.6784(6)	0.4956(5)	0.2791(4)	0.064(2)
N9	0.4416(8)	0.9478(4)	0.3669(4)	0.088(2)
C81	0.9889(9)	0.1603(6)	0.3572(4)	0.079(3)
C82	0.9073(11)	0.1148(6)	0.3270(5)	0.093(3)
F71	0.5567(9)	0.9737(5)	0.1424(5)	0.167(4)
F72	0.6894(13)	0.0418(5)	0.1746(5)	0.231(6)
F73	0.7262(11)	0.9654(6)	0.0855(4)	0.178(5)
C74	0.6835(17)	0.9730(8)	0.1449(8)	0.127(5)

Table S5. Bond lengths (Å) for 4mer-(NHTFSI)₄.

Si1-O5	1.609(4)	Si1-O1	1.611(4)
Si1-O2	1.613(4)	Si1-C11	1.850(6)
Si2-O8	1.604(4)	Si2-O3	1.613(4)
Si2-O4	1.619(4)	Si2-C21	1.841(6)
Si3-O6	1.606(4)	Si3-O3	1.611(4)
Si3-O2	1.612(4)	Si3-C31	1.854(6)
Si4-O7	1.609(4)	Si4-O4	1.610(4)
Si4-O1	1.622(4)	Si4-C41	1.839(6)
Si5-O6	1.651(4)	Si5-C7A	1.847(7)
Si5-C8A	1.851(6)	Si5-C51	1.870(7)
S8-O82	1.390(6)	S8-O81	1.414(5)
S8-N8	1.579(7)	S8-C84	1.820(8)
Si7-O5	1.649(4)	Si7-C2A	1.836(8)
Si7-C1A	1.848(8)	Si7-C71	1.870(8)
Si8-O7	1.639(4)	Si8-C3A	1.845(8)
Si8-C4A	1.848(8)	Si8-C81	1.886(10)
Si9-O8	1.659(4)	Si9-C5A	1.853(8)
Si9-C91	1.856(8)	Si9-C6A	1.861(9)

C11-C12	1.390(9)	C11-C16	1.400(9)
C12-C13	1.389(9)	C16-C15	1.395(10)
C14-C13	1.370(11)	C14-C15	1.373(12)
C21-C26	1.386(8)	C21-C22	1.402(9)
C25-C24	1.367(9)	C25-C26	1.371(8)
C23-C24	1.353(10)	C23-C22	1.390(10)
C31-C36	1.370(9)	C31-C32	1.386(8)
C32-C33	1.396(9)	C34-C33	1.365(10)
C34-C35	1.366(10)	C36-C35	1.394(9)
C41-C46	1.360(9)	C41-C42	1.394(9)
C42-C83A	1.375(10)	C44-C83A	1.336(11)
C44-C45	1.360(11)	C46-C45	1.413(10)
C84-F81	1.314(7)	C84-F83	1.320(8)
C84-F82	1.326(8)	C83-N8	1.456(10)
C83-C82	1.556(12)	C71-C72	1.499(11)
C72-C73	1.558(13)	C73-N7	1.427(8)
N7-S7	1.683(10)	S7-O71	1.386(6)
S7-O72	1.389(7)	S7-C74	1.787(14)
C51-C52	1.520(9)	C53-N5	1.441(9)
C53-C52	1.514(9)	C54-F52	1.300(10)
C54-F51	1.309(10)	C54-F53	1.336(10)
C54-S5	1.827(9)	N5-S5	1.584(6)
S5-O51	1.391(6)	S5-O52	1.420(6)
C91-C92	1.472(11)	C92-C93	1.547(12)
C94-F93	1.285(8)	C94-F91	1.329(9)
C94-F92	1.339(9)	C94-S9	1.836(8)
C93-N9	1.403(12)	S9-O91	1.370(7)
S9-O92	1.398(6)	S9-N9	1.646(8)
C81-C82	1.469(12)	F71-C74	1.444(17)
F72-C74	1.300(13)	F73-C74	1.214(17)

Table S6. Bond angles (°) for 4mer-(NHTFSI)₄.

O5-Si1-O1	110.4(2)	O5-Si1-O2	106.8(2)
O1-Si1-O2	110.2(2)	O5-Si1-C11	111.7(2)
O1-Si1-C11	109.1(3)	O2-Si1-C11	108.6(2)
O8-Si2-O3	109.8(2)	O8-Si2-O4	106.8(2)
O3-Si2-O4	110.7(2)	O8-Si2-C21	111.6(2)
O3-Si2-C21	108.8(2)	O4-Si2-C21	109.1(2)
O6-Si3-O3	108.7(2)	O6-Si3-O2	109.3(2)
O3-Si3-O2	110.4(2)	O6-Si3-C31	110.9(2)

O3-Si3-C31	108.1(2)	O2-Si3-C31	109.4(2)
O7-Si4-O4	109.0(2)	O7-Si4-O1	109.5(2)
O4-Si4-O1	110.1(2)	O7-Si4-C41	109.7(2)
O4-Si4-C41	109.9(2)	O1-Si4-C41	108.6(2)
O6-Si5-C7A	109.6(3)	O6-Si5-C8A	108.7(3)
C7A-Si5-C8A	109.7(3)	O6-Si5-C51	109.8(3)
C7A-Si5-C51	110.6(3)	C8A-Si5-C51	108.5(3)
O82-S8-O81	122.1(4)	O82-S8-N8	112.0(4)
O81-S8-N8	108.7(3)	O82-S8-C84	103.7(3)
O81-S8-C84	102.8(3)	N8-S8-C84	105.8(3)
O5-Si7-C2A	108.1(3)	O5-Si7-C1A	108.1(3)
C2A-Si7-C1A	112.0(4)	O5-Si7-C71	106.8(4)
C2A-Si7-C71	108.0(4)	C1A-Si7-C71	113.5(4)
O7-Si8-C3A	108.6(3)	O7-Si8-C4A	107.8(3)
C3A-Si8-C4A	109.4(4)	O7-Si8-C81	108.5(3)
C3A-Si8-C81	107.6(4)	C4A-Si8-C81	114.8(4)
O8-Si9-C5A	107.5(3)	O8-Si9-C91	105.6(3)
C5A-Si9-C91	112.6(4)	O8-Si9-C6A	109.8(3)
C5A-Si9-C6A	111.4(4)	C91-Si9-C6A	109.7(4)
C12-C11-C16	118.1(6)	C12-C11-Si1	121.8(5)
C16-C11-Si1	120.1(5)	C13-C12-C11	120.2(7)
C15-C16-C11	121.9(7)	C13-C14-C15	121.8(7)
C14-C13-C12	120.1(7)	C14-C15-C16	117.9(7)
C26-C21-C22	115.8(6)	C26-C21-Si2	123.3(4)
C22-C21-Si2	120.9(5)	C24-C25-C26	120.7(6)
C25-C26-C21	122.0(6)	C24-C23-C22	120.0(7)
C23-C24-C25	119.7(6)	C23-C22-C21	121.7(6)
C36-C31-C32	116.8(6)	C36-C31-Si3	123.4(5)
C32-C31-Si3	119.8(5)	C31-C32-C33	121.7(6)
C33-C34-C35	119.8(7)	C34-C33-C32	119.8(6)
C31-C36-C35	122.2(6)	C34-C35-C36	119.7(7)
C46-C41-C42	116.8(6)	C46-C41-Si4	124.1(5)
C42-C41-Si4	119.1(5)	C83A-C42-C41	121.1(7)
C83A-C44-C45	120.5(7)	C41-C46-C45	121.9(7)
C44-C45-C46	118.6(8)	C44-C83A-C42	121.1(7)
Si3-O3-Si2	161.7(3)	Si3-O2-Si1	153.5(3)
Si1-O1-Si4	155.5(3)	Si4-O4-Si2	152.3(3)
Si1-O5-Si7	144.6(3)	Si3-O6-Si5	144.7(3)
Si4-O7-Si8	142.0(3)	Si2-O8-Si9	137.6(3)
F81-C84-F83	108.2(6)	F81-C84-F82	108.1(6)
F83-C84-F82	109.1(6)	F81-C84-S8	110.0(5)

F83-C84-S8	111.0(5)	F82-C84-S8	110.3(5)
N8-C83-C82	111.5(7)	C83-N8-S8	123.7(6)
C72-C71-Si7	116.0(6)	C71-C72-C73	112.8(8)
N7-C73-C72	107.1(8)	C73-N7-S7	120.8(8)
O71-S7-O72	120.6(5)	O71-S7-N7	113.8(5)
O72-S7-N7	102.5(6)	O71-S7-C74	109.3(7)
O72-S7-C74	103.1(7)	N7-S7-C74	106.2(5)
C52-C51-Si5	117.1(5)	N5-C53-C52	114.0(6)
C53-C52-C51	113.8(5)	F52-C54-F51	109.0(8)
F52-C54-F53	106.8(7)	F51-C54-F53	109.4(7)
F52-C54-S5	111.2(6)	F51-C54-S5	111.2(6)
F53-C54-S5	109.0(7)	C53-N5-S5	124.9(5)
O51-S5-O52	123.1(4)	O51-S5-N5	110.7(3)
O52-S5-N5	108.9(4)	O51-S5-C54	104.6(5)
O52-S5-C54	101.7(4)	N5-S5-C54	106.1(4)
C92-C91-Si9	111.4(6)	C91-C92-C93	114.8(7)
F93-C94-F91	109.6(7)	F93-C94-F92	107.1(7)
F91-C94-F92	107.1(6)	F93-C94-S9	113.7(5)
F91-C94-S9	110.4(6)	F92-C94-S9	108.7(5)
N9-C93-C92	112.1(8)	O91-S9-O92	120.4(4)
O91-S9-N9	116.1(4)	O92-S9-N9	105.4(4)
O91-S9-C94	103.3(4)	O92-S9-C94	105.1(4)
N9-S9-C94	105.0(4)	C93-N9-S9	121.1(8)
C82-C81-Si8	116.4(7)	C81-C82-C83	114.5(9)
F73-C74-F72	114.6(14)	F73-C74-F71	105.8(12)
F72-C74-F71	105.8(14)	F73-C74-S7	118.1(12)
F72-C74-S7	109.4(10)	F71-C74-S7	101.4(10)

Table S7. Torsion angles (°) for 4mer-(NHTFSI)₄.

O5-Si1-C11-C12	-107.6(5)	O1-Si1-C11-C12	14.7(6)
O2-Si1-C11-C12	134.8(5)	O5-Si1-C11-C16	72.9(5)
O1-Si1-C11-C16	-164.8(5)	O2-Si1-C11-C16	-44.6(6)
C16-C11-C12-C13	-2.1(9)	Si1-C11-C12-C13	178.4(5)
C12-C11-C16-C15	0.7(10)	Si1-C11-C16-C15	-179.8(5)
C15-C14-C13-C12	-1.3(11)	C11-C12-C13-C14	2.5(10)
C13-C14-C15-C16	-0.2(12)	C11-C16-C15-C14	0.5(11)
O8-Si2-C21-C26	-118.0(5)	O3-Si2-C21-C26	3.2(5)
O4-Si2-C21-C26	124.2(5)	O8-Si2-C21-C22	62.7(5)
O3-Si2-C21-C22	-176.0(5)	O4-Si2-C21-C22	-55.1(5)
C24-C25-C26-C21	1.2(9)	C22-C21-C26-C25	-1.8(8)

Si2-C21-C26-C25	178.9(5)	C22-C23-C24-C25	-2.1(11)
C26-C25-C24-C23	0.8(10)	C24-C23-C22-C21	1.5(12)
C26-C21-C22-C23	0.5(10)	Si2-C21-C22-C23	179.8(6)
O6-Si3-C31-C36	-108.6(5)	O3-Si3-C31-C36	132.3(5)
O2-Si3-C31-C36	12.1(6)	O6-Si3-C31-C32	71.1(5)
O3-Si3-C31-C32	-48.0(5)	O2-Si3-C31-C32	-168.3(5)
C36-C31-C32-C33	0.8(9)	Si3-C31-C32-C33	-178.9(5)
C35-C34-C33-C32	-2.0(11)	C31-C32-C33-C34	1.2(11)
C32-C31-C36-C35	-2.0(10)	Si3-C31-C36-C35	177.6(5)
C33-C34-C35-C36	0.8(11)	C31-C36-C35-C34	1.3(11)
O7-Si4-C41-C46	-113.9(6)	O4-Si4-C41-C46	5.9(6)
O1-Si4-C41-C46	126.4(6)	O7-Si4-C41-C42	65.8(6)
O4-Si4-C41-C42	-174.4(5)	O1-Si4-C41-C42	-53.9(6)
C46-C41-C42-C83A	2.2(11)	Si4-C41-C42-C83A	-177.6(6)
C42-C41-C46-C45	-2.1(11)	Si4-C41-C46-C45	177.6(6)
C83A-C44-C45-C46	2.1(14)	C41-C46-C45-C44	0.0(13)
C45-C44-C83A-C42	-2.1(14)	C41-C42-C83A-C44	-0.1(13)
O6-Si3-O3-Si2	82.2(9)	O2-Si3-O3-Si2	-37.7(10)
C31-Si3-O3-Si2	-157.3(9)	O8-Si2-O3-Si3	-92.6(9)
O4-Si2-O3-Si3	25.0(10)	C21-Si2-O3-Si3	145.0(9)
O6-Si3-O2-Si1	-114.3(6)	O3-Si3-O2-Si1	5.3(7)
C31-Si3-O2-Si1	124.1(6)	O5-Si1-O2-Si3	119.0(6)
O1-Si1-O2-Si3	-0.9(7)	C11-Si1-O2-Si3	-120.3(6)
O5-Si1-O1-Si4	-64.7(7)	O2-Si1-O1-Si4	53.1(7)
C11-Si1-O1-Si4	172.2(6)	O7-Si4-O1-Si1	57.3(7)
O4-Si4-O1-Si1	-62.6(7)	C41-Si4-O1-Si1	177.1(6)
O7-Si4-O4-Si2	-115.9(6)	O1-Si4-O4-Si2	4.3(7)
C41-Si4-O4-Si2	123.8(6)	O8-Si2-O4-Si4	131.1(6)
O3-Si2-O4-Si4	11.6(7)	C21-Si2-O4-Si4	-108.1(6)
O1-Si1-O5-Si7	-60.1(5)	O2-Si1-O5-Si7	-179.9(4)
C11-Si1-O5-Si7	61.5(5)	C2A-Si7-O5-Si1	155.4(5)
C1A-Si7-O5-Si1	34.0(6)	C71-Si7-O5-Si1	-88.6(5)
O3-Si3-O6-Si5	-150.4(4)	O2-Si3-O6-Si5	-29.8(5)
C31-Si3-O6-Si5	90.9(5)	C7A-Si5-O6-Si3	78.4(5)
C8A-Si5-O6-Si3	-161.8(5)	C51-Si5-O6-Si3	-43.2(5)
O4-Si4-O7-Si8	-8.9(5)	O1-Si4-O7-Si8	-129.4(4)
C41-Si4-O7-Si8	111.5(5)	C3A-Si8-O7-Si4	63.5(6)
C4A-Si8-O7-Si4	-178.1(5)	C81-Si8-O7-Si4	-53.2(6)
O3-Si2-O8-Si9	-44.7(5)	O4-Si2-O8-Si9	-164.8(4)
C21-Si2-O8-Si9	76.0(5)	C5A-Si9-O8-Si2	100.9(5)
C91-Si9-O8-Si2	-138.6(5)	C6A-Si9-O8-Si2	-20.4(6)

O82-S8-C84-F81	175.2(5)	O81-S8-C84-F81	-56.7(6)
N8-S8-C84-F81	57.2(6)	O82-S8-C84-F83	55.5(6)
O81-S8-C84-F83	-176.4(5)	N8-S8-C84-F83	-62.5(5)
O82-S8-C84-F82	-65.6(6)	O81-S8-C84-F82	62.5(5)
N8-S8-C84-F82	176.4(5)	C82-C83-N8-S8	91.9(9)
O82-S8-N8-C83	-18.4(7)	O81-S8-N8-C83	-156.3(6)
C84-S8-N8-C83	93.9(6)	O5-Si7-C71-C72	80.4(8)
C2A-Si7-C71-C72	-163.5(7)	C1A-Si7-C71-C72	-38.7(9)
Si7-C71-C72-C73	-173.6(7)	C71-C72-C73-N7	67.4(11)
C72-C73-N7-S7	-121.1(8)	C73-N7-S7-O71	23.9(9)
C73-N7-S7-O72	155.8(8)	C73-N7-S7-C74	-96.4(10)
O6-Si5-C51-C52	65.3(6)	C7A-Si5-C51-C52	-55.7(6)
C8A-Si5-C51-C52	-176.1(5)	N5-C53-C52-C51	62.9(8)
Si5-C51-C52-C53	175.3(5)	C52-C53-N5-S5	111.5(6)
C53-N5-S5-O51	-28.2(7)	C53-N5-S5-O52	-166.5(6)
C53-N5-S5-C54	84.7(7)	F52-C54-S5-O51	53.3(8)
F51-C54-S5-O51	-68.4(8)	F53-C54-S5-O51	170.8(6)
F52-C54-S5-O52	-177.6(7)	F51-C54-S5-O52	60.7(8)
F53-C54-S5-O52	-60.1(7)	F52-C54-S5-N5	-63.8(8)
F51-C54-S5-N5	174.5(6)	F53-C54-S5-N5	53.7(7)
O8-Si9-C91-C92	-58.6(6)	C5A-Si9-C91-C92	58.5(7)
C6A-Si9-C91-C92	-176.8(6)	Si9-C91-C92-C93	-175.8(6)
C91-C92-C93-N9	-63.8(11)	F93-C94-S9-O91	71.3(7)
F91-C94-S9-O91	-52.3(6)	F92-C94-S9-O91	-169.5(6)
F93-C94-S9-O92	-55.7(7)	F91-C94-S9-O92	-179.4(6)
F92-C94-S9-O92	63.5(6)	F93-C94-S9-N9	-166.7(6)
F91-C94-S9-N9	69.7(7)	F92-C94-S9-N9	-47.4(7)
C92-C93-N9-S9	-130.0(7)	O91-S9-N9-C93	41.6(8)
O92-S9-N9-C93	177.6(7)	C94-S9-N9-C93	-71.7(8)
O7-Si8-C81-C82	-62.0(7)	C3A-Si8-C81-C82	-179.4(7)
C4A-Si8-C81-C82	58.6(8)	Si8-C81-C82-C83	-175.1(6)
N8-C83-C82-C81	63.0(12)	O71-S7-C74-F73	-170.5(12)
O72-S7-C74-F73	60.0(14)	N7-S7-C74-F73	-47.4(14)
O71-S7-C74-F72	55.9(15)	O72-S7-C74-F72	-73.5(14)
N7-S7-C74-F72	179.0(12)	O71-S7-C74-F71	-55.5(9)
O72-S7-C74-F71	175.0(8)	N7-S7-C74-F71	67.6(10)

Table S8. Anisotropic atomic displacement parameters ($\text{\AA}^2$) for 4mer-(NHTFSI)₄.

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2 [h^2 a^{*2} U_{11} + \dots + 2 h k a^* b^* U_{12}]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Si1	0.0191(8)	0.0298(9)	0.0279(9)	0.0078(7)	-0.0041(6)	-0.0043(6)
Si2	0.0198(8)	0.0289(9)	0.0303(9)	0.0040(7)	-0.0084(7)	-0.0042(6)
Si3	0.0198(8)	0.0283(9)	0.0291(9)	-0.0009(7)	-0.0037(6)	-0.0056(6)
Si4	0.0203(8)	0.0326(9)	0.0265(9)	0.0073(7)	-0.0043(6)	-0.0082(7)
Si5	0.0293(9)	0.0371(10)	0.0296(10)	-0.0013(7)	-0.0052(7)	-0.0042(7)
S8	0.0544(12)	0.0515(11)	0.0466(12)	-0.0012(9)	-0.0092(9)	0.0032(9)
Si7	0.0294(10)	0.0525(12)	0.0435(12)	0.0019(9)	0.0007(8)	0.0082(8)
Si8	0.0421(10)	0.0498(11)	0.0279(10)	0.0078(8)	-0.0043(8)	-0.0168(9)
Si9	0.0445(11)	0.0442(11)	0.0568(13)	0.0072(9)	-0.0315(10)	-0.0021(9)
C11	0.022(3)	0.044(4)	0.033(4)	0.012(3)	-0.002(3)	-0.005(3)
C12	0.019(3)	0.068(5)	0.035(4)	0.009(3)	0.000(3)	-0.005(3)
C16	0.033(4)	0.058(5)	0.056(5)	0.030(4)	-0.016(3)	-0.010(3)
C14	0.027(4)	0.117(7)	0.051(5)	0.043(5)	-0.002(3)	-0.002(4)
C13	0.022(3)	0.108(7)	0.036(4)	0.012(4)	-0.004(3)	-0.012(4)
C15	0.039(4)	0.078(6)	0.075(6)	0.048(5)	-0.013(4)	-0.014(4)
C21	0.021(3)	0.029(3)	0.033(3)	0.005(3)	-0.008(2)	-0.002(2)
C25	0.032(3)	0.046(4)	0.038(4)	-0.005(3)	0.001(3)	-0.014(3)
C26	0.028(3)	0.035(3)	0.034(4)	-0.004(3)	-0.009(3)	-0.012(3)
C23	0.072(5)	0.032(4)	0.070(6)	-0.014(4)	0.006(4)	-0.009(4)
C24	0.041(4)	0.054(5)	0.046(4)	-0.012(4)	0.005(3)	-0.007(3)
C22	0.052(4)	0.035(4)	0.063(5)	0.002(4)	0.015(4)	-0.011(3)
C31	0.029(3)	0.031(3)	0.036(4)	-0.009(3)	-0.003(3)	-0.016(3)
C32	0.025(3)	0.053(4)	0.057(5)	0.009(3)	-0.009(3)	-0.017(3)
C34	0.050(5)	0.054(5)	0.061(5)	-0.005(4)	0.017(4)	-0.022(4)
C33	0.023(3)	0.065(5)	0.072(5)	-0.001(4)	0.001(3)	-0.015(3)
C36	0.039(4)	0.041(4)	0.046(4)	0.000(3)	0.011(3)	-0.005(3)
C35	0.052(5)	0.047(4)	0.053(5)	0.006(4)	0.017(4)	-0.011(3)
C41	0.031(3)	0.038(4)	0.026(3)	0.011(3)	-0.005(3)	-0.014(3)
C42	0.039(4)	0.065(5)	0.074(6)	-0.011(4)	-0.015(4)	-0.012(4)
C44	0.065(5)	0.082(6)	0.053(5)	0.002(4)	-0.028(4)	-0.036(5)
C46	0.049(4)	0.060(5)	0.055(5)	-0.003(4)	-0.023(4)	-0.007(4)
C45	0.081(6)	0.068(6)	0.073(6)	-0.022(5)	-0.026(5)	-0.011(5)
C83A	0.045(5)	0.089(7)	0.085(6)	-0.013(5)	-0.028(4)	-0.025(5)
O3	0.020(2)	0.031(2)	0.044(3)	-0.0038(19)	-0.0025(18)	-0.0052(17)
O2	0.020(2)	0.029(2)	0.038(2)	0.0049(18)	-0.0074(17)	-0.0058(16)
O1	0.024(2)	0.038(2)	0.037(2)	0.0069(19)	-0.0093(18)	-0.0118(18)
O4	0.019(2)	0.035(2)	0.034(2)	0.0057(18)	-0.0048(17)	-0.0080(17)
O5	0.024(2)	0.041(2)	0.033(2)	0.0037(19)	-0.0028(17)	-0.0008(18)
O6	0.031(2)	0.031(2)	0.033(2)	-0.0016(18)	-0.0101(18)	0.0012(17)
O7	0.022(2)	0.048(3)	0.031(2)	0.0081(19)	-0.0016(17)	-0.0065(18)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O8	0.037(2)	0.047(3)	0.033(2)	0.006(2)	-0.0147(19)	-0.005(2)
C84	0.033(4)	0.066(5)	0.044(5)	0.000(4)	-0.011(3)	-0.007(3)
C83	0.133(8)	0.056(5)	0.044(5)	0.003(4)	-0.022(5)	-0.017(5)
N8	0.072(4)	0.059(4)	0.053(4)	0.008(3)	-0.015(3)	-0.015(3)
O81	0.080(4)	0.076(4)	0.042(3)	-0.010(3)	-0.005(3)	-0.015(3)
O82	0.079(4)	0.085(4)	0.064(4)	-0.001(3)	-0.021(3)	0.016(3)
F81	0.044(2)	0.059(3)	0.062(3)	0.009(2)	-0.009(2)	0.0006(19)
F83	0.072(3)	0.073(3)	0.062(3)	-0.020(2)	-0.020(2)	-0.004(2)
F82	0.054(3)	0.076(3)	0.099(4)	0.006(3)	0.004(3)	-0.028(2)
C71	0.080(6)	0.075(6)	0.068(6)	-0.013(5)	-0.024(5)	0.031(5)
C72	0.079(6)	0.065(6)	0.089(7)	-0.019(5)	-0.041(5)	0.010(5)
C73	0.074(7)	0.170(12)	0.075(7)	0.019(7)	-0.023(6)	0.015(7)
N7	0.085(6)	0.134(8)	0.068(6)	0.010(5)	-0.001(5)	0.011(5)
S7	0.0743(16)	0.117(2)	0.0610(16)	0.0114(14)	-0.0222(12)	-0.0298(14)
O71	0.092(5)	0.220(9)	0.049(4)	-0.026(5)	0.018(4)	-0.048(5)
O72	0.073(5)	0.216(10)	0.139(8)	0.022(7)	-0.034(5)	-0.028(5)
C51	0.045(4)	0.041(4)	0.045(4)	-0.012(3)	-0.004(3)	-0.005(3)
C53	0.053(4)	0.045(4)	0.041(4)	-0.004(3)	-0.007(3)	-0.003(3)
C52	0.046(4)	0.038(4)	0.048(4)	-0.003(3)	-0.009(3)	-0.005(3)
C54	0.089(7)	0.053(5)	0.062(6)	-0.005(5)	0.019(5)	-0.024(5)
N5	0.049(4)	0.067(4)	0.042(4)	-0.002(3)	-0.004(3)	-0.006(3)
S5	0.0892(16)	0.0474(11)	0.0461(12)	0.0017(9)	0.0064(11)	-0.0065(10)
O52	0.100(5)	0.089(5)	0.078(5)	0.002(4)	0.044(4)	0.000(4)
O51	0.136(6)	0.098(5)	0.042(3)	0.015(3)	-0.039(3)	-0.064(4)
F53	0.138(5)	0.075(4)	0.109(5)	-0.007(3)	0.012(4)	-0.050(4)
F52	0.096(4)	0.066(3)	0.104(4)	0.004(3)	0.017(3)	0.012(3)
F51	0.146(5)	0.067(3)	0.088(4)	0.038(3)	0.004(4)	-0.008(3)
C91	0.062(5)	0.058(5)	0.072(6)	0.003(4)	-0.031(4)	0.002(4)
C92	0.062(5)	0.058(5)	0.081(6)	0.008(4)	-0.023(5)	-0.004(4)
C94	0.071(6)	0.058(5)	0.058(6)	0.006(4)	-0.022(4)	-0.028(4)
C93	0.087(7)	0.082(7)	0.093(8)	0.009(6)	-0.023(6)	-0.003(5)
S9	0.0775(16)	0.0532(12)	0.0667(15)	-0.0060(11)	-0.0338(12)	-0.0055(11)
O92	0.079(4)	0.067(4)	0.056(4)	-0.006(3)	-0.026(3)	-0.017(3)
O91	0.095(5)	0.102(5)	0.071(4)	-0.037(4)	-0.030(4)	0.020(4)
F93	0.106(4)	0.058(3)	0.082(4)	0.011(2)	-0.034(3)	-0.029(3)
F92	0.080(4)	0.100(4)	0.103(4)	0.030(3)	-0.049(3)	-0.028(3)
F91	0.141(5)	0.087(4)	0.055(3)	-0.008(3)	-0.036(3)	-0.009(3)
C8A	0.045(4)	0.050(4)	0.043(4)	-0.005(3)	-0.013(3)	-0.008(3)
C5A	0.074(5)	0.036(4)	0.074(6)	-0.001(4)	-0.045(4)	-0.003(4)
C7A	0.045(4)	0.068(5)	0.043(4)	-0.001(4)	0.002(3)	-0.017(4)

	U₁₁	U₂₂	U₃₃	U₂₃	U₁₃	U₁₂
C6A	0.045(5)	0.102(7)	0.103(8)	0.023(6)	-0.034(5)	-0.020(5)
C3A	0.061(5)	0.122(8)	0.041(5)	0.004(5)	-0.008(4)	-0.041(5)
C4A	0.050(5)	0.149(9)	0.037(5)	0.009(5)	0.007(4)	-0.033(5)
C2A	0.041(4)	0.096(6)	0.055(5)	-0.012(5)	0.011(4)	0.000(4)
C1A	0.033(4)	0.095(6)	0.061(5)	-0.001(5)	0.006(4)	-0.010(4)
N9	0.140(7)	0.055(4)	0.064(5)	0.002(4)	-0.032(5)	0.000(4)
C81	0.101(7)	0.097(7)	0.050(5)	0.040(5)	-0.030(5)	-0.038(6)
C82	0.148(10)	0.085(7)	0.063(6)	0.033(5)	-0.042(6)	-0.055(7)
F71	0.183(8)	0.132(6)	0.167(8)	-0.019(5)	-0.098(7)	0.047(6)
F72	0.50(2)	0.102(6)	0.126(7)	0.005(5)	-0.065(9)	-0.131(9)
F73	0.331(14)	0.186(8)	0.050(4)	0.019(5)	-0.013(6)	-0.134(9)
C74	0.181(15)	0.085(9)	0.124(13)	-0.012(8)	-0.062(11)	-0.028(9)

Table S9. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($\text{\AA}^2$) for 4mer-(NHTFSI)₄.

	x/a	y/b	z/c	U(eq)
H12	-0.1405	0.4761	0.0772	0.049
H16	0.0124	0.6608	0.1373	0.058
H14	-0.1432	0.6748	-0.0422	0.08
H13	-0.1979	0.5502	-0.0213	0.067
H15	-0.0382	0.7322	0.0359	0.076
H25	0.5016	0.2476	-0.0083	0.046
H26	0.4077	0.3183	0.0891	0.037
H23	0.3748	0.0491	0.0515	0.071
H24	0.4876	0.1128	-0.0269	0.057
H22	0.2853	0.1174	0.1526	0.061
H32	0.5080	0.4304	0.1711	0.052
H34	0.5639	0.5959	0.0263	0.066
H33	0.6366	0.4803	0.0894	0.063
H36	0.2320	0.6039	0.1188	0.052
H35	0.3610	0.6570	0.0401	0.062
H42	-0.2473	0.3438	0.1588	0.07
H44	-0.2441	0.1545	0.0330	0.075
H46	0.0536	0.1712	0.1272	0.065
H45	-0.0441	0.1022	0.0506	0.088
H83A	-0.3414	0.2751	0.0838	0.084
H83B	-0.1051	-0.0056	0.3011	0.093
H83C	0.0316	0.0061	0.2988	0.093
H8	0.0299	-0.0157	0.4136	0.072

	x/a	y/b	z/c	U(eq)
H71A	-0.3727	0.7054	0.3023	0.096
H71B	-0.2432	0.7135	0.2685	0.096
H72A	-0.4205	0.6539	0.2002	0.095
H72B	-0.2871	0.6512	0.1658	0.095
H73A	-0.4311	0.7702	0.1285	0.133
H73B	-0.4493	0.8044	0.2067	0.133
H7	-0.2263	0.7799	0.1315	0.121
H51A	0.2314	0.7036	0.3675	0.053
H51B	0.3163	0.6691	0.3005	0.053
H53A	0.0518	0.8005	0.2363	0.056
H53B	0.1122	0.8191	0.3044	0.056
H52A	0.1489	0.6610	0.2406	0.053
H52B	0.0607	0.6884	0.3082	0.053
H5	0.2947	0.7979	0.2399	0.064
H91A	0.5038	0.0853	0.3056	0.077
H91B	0.5591	0.1130	0.3727	0.077
H92A	0.3166	0.1001	0.3647	0.081
H92B	0.3657	0.1343	0.4311	0.081
H93A	0.4919	0.0018	0.4460	0.106
H93B	0.3521	-0.0025	0.4520	0.106
H8AA	0.3911	0.4838	0.4364	0.068
H8AB	0.3614	0.5788	0.4581	0.068
H8AC	0.4549	0.5475	0.3933	0.068
H5AA	0.3714	0.3561	0.3808	0.091
H5AB	0.4784	0.3020	0.4198	0.091
H5AC	0.3461	0.2822	0.4290	0.091
H7AA	0.0509	0.5493	0.3787	0.077
H7AB	0.0906	0.5893	0.4457	0.077
H7AC	0.1274	0.4923	0.4328	0.077
H6AA	0.5702	0.2265	0.2183	0.121
H6AB	0.6420	0.2415	0.2831	0.121
H6AC	0.5475	0.3156	0.2535	0.121
H3AA	0.1301	0.2967	0.3786	0.107
H3AB	0.0593	0.2983	0.4533	0.107
H3AC	0.0329	0.3766	0.4044	0.107
H4AA	-0.2303	0.3659	0.4254	0.116
H4AB	-0.1998	0.2828	0.4680	0.116
H4AC	-0.2733	0.2856	0.4009	0.116
H2AA	-0.1915	0.5310	0.4129	0.1
H2AB	-0.3130	0.5995	0.4178	0.1

	x/a	y/b	z/c	U(eq)
H2AC	-0.1854	0.6259	0.4049	0.1
H1AA	-0.3161	0.4887	0.2288	0.096
H1AB	-0.4064	0.5120	0.2964	0.096
H1AC	-0.2860	0.4436	0.3001	0.096
H9	0.4920	-0.0471	0.3311	0.105
H81A	0.0659	0.1504	0.3280	0.095
H81B	0.0064	0.1372	0.4032	0.095
H82A	-0.1045	0.1339	0.2792	0.111
H82B	-0.1721	0.1280	0.3537	0.111