Electronic Supporting Information For:

Design principles and multiscale simulations for determining nanostructure in self-assembling ionic liquids

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Table S1: Comparison force field parameters created by Liu, Huang, and Wang[1] to EFP and SAPT2+3 energies[2] for various imidazolium cations with the chloride anion. The EFP energy shown here is that obtained with the aug-cc-pVDZ basis set. Structures and energies for EFP and SAPT 2+3 can be found in reference 2. Structure abbreviations are the same as used in reference 2: dmim: dimethyl-imidazolium, emim: 1-methyl-3-ethyl-imidazolium, bmim: 1-methyl-3-butyl-imidazolium.

Ion	Conf	Force Field Energy (kJ/mol)	EFP Energy (kJ/mol)	SAPT 2+3 Energy (kJ/mol)	Force Field Error (%)	EFP Error (%)
dmim	1	364.3	414.1	423.4	14.0	2.2
dmim	2	381.2	437.7	405.9	6.1	7.8
emim	1	380.9	388.9	413.4	7.9	5.9
emim	2	396.4	408.6	409.7	3.3	0.3
emim	3	392.5	405.6	405.4	3.2	0.0
emim	4	382.0	406.4	418.2	8.6	2.8
bmim	1	375.0	383.4	411.6	8.9	6.9
bmim	2	391.6	380.4	408.1	4.0	6.8
bmim	3	387.3	410.6	406.5	4.7	1.0
bmim	4	377.1	411.6	419.3	10.1	1.8
				Average	7.1	3.6

1. Z. P. Liu, S. P. Huang and W. C. Wang, *J Phys Chem B*, 2004, **108**, 12978-12989.
2. S. Y. S. Tan and E. I. Izgorodina, *Journal of Chemical Theory and Computation*, 2016, **12**, 2553-2568.

Table S2: Table of simulated and experimental water self-diffusion coefficients. A temperature of 310 K was used for the production molecular dynamics simulations because the simulated value at that temperature matched the experimental self-diffusion coefficient at room temperature.

Temperature (K)	Simulated Self Diffusion (Å^2/ps)Simulated	Experimental Self Diffusion (Å^2/ps)
288	-	0.176
298	-	0.230
300	0.170	-
310	0.202	-
323	-	0.398
325	0.430	-
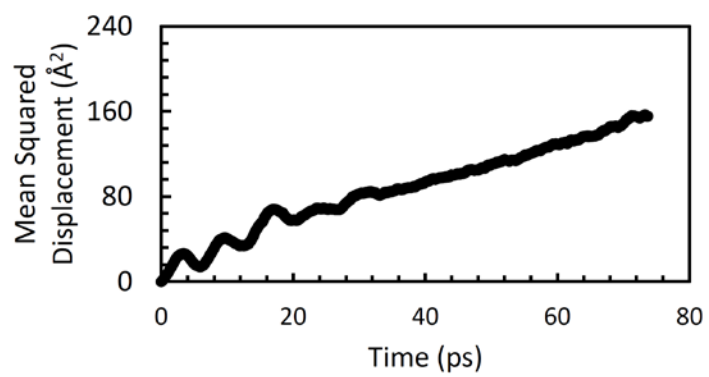
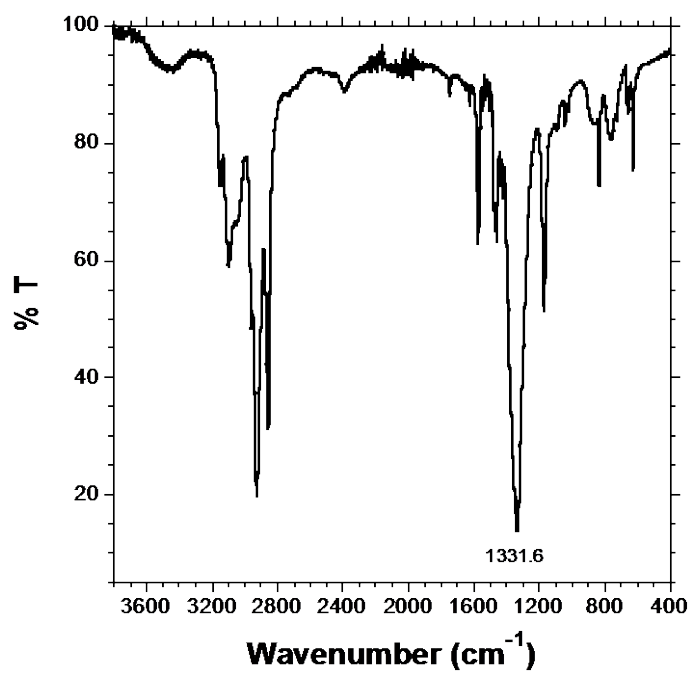**Figure S1.** Mean squared displacement (MSD) *vs.* time for MD simulation

Figure S2A. ATR/FT-IR of dried 1-decyl-3-methylimidazolium nitrate, $C_{10}mim^+NO_3^-$, but still contains a residual 0.92% (w/w) H_2O as determined by TGA.
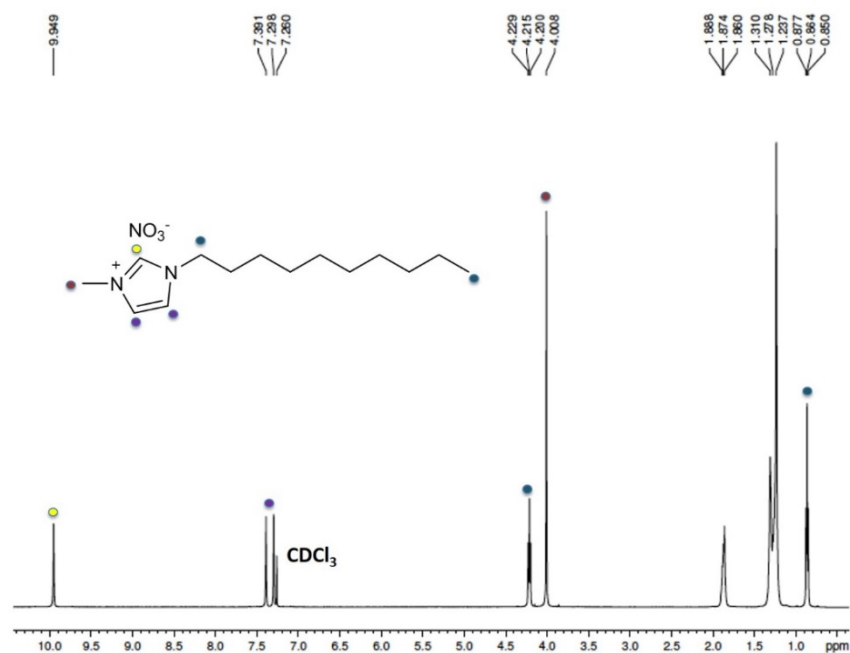
Figure S2B. ^1H NMR of dried 1-decyl-3-methylimidazolium nitrate, $C_{10}mim^+NO_3^-$ in $CDCl_3$. (Sample contains a residual 0.92% (w/w) H_2O, as determined by TGA).
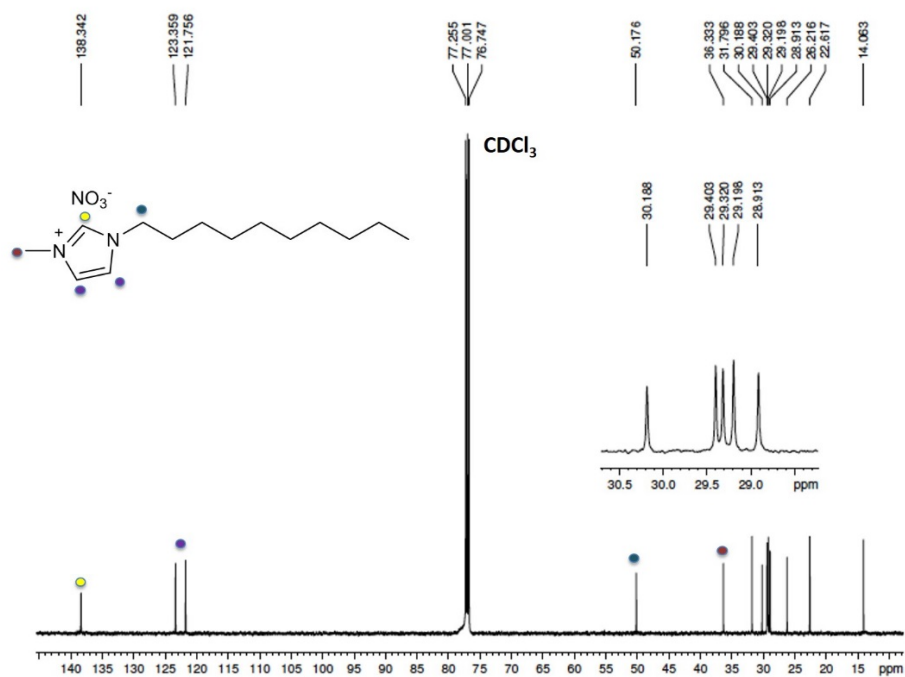

Figure S2C. ^{13}C NMR of dried 1-decyl-3-methylimidazolium nitrate, $C_{10}mim^+NO_3^-$ in $CDCl_3$. (Sample contains a residual 0.92% (w/w) H_2O as determined by TGA.)
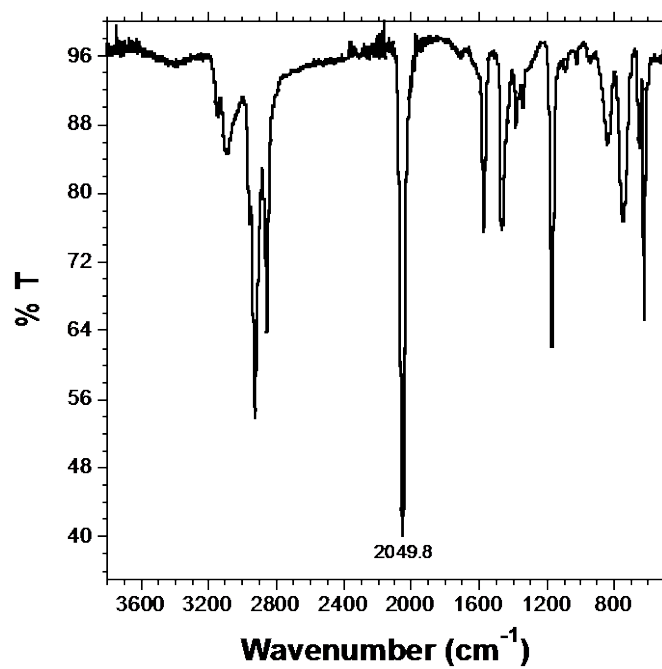
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Figure S3A. ATR/FT-IR of dried -decyl-3-methylimidazolium thiocyanate, $C_{10}mim^+SCN^-$, but still contains a residual 4.9% (w/w) H_2O as determined by TGA.
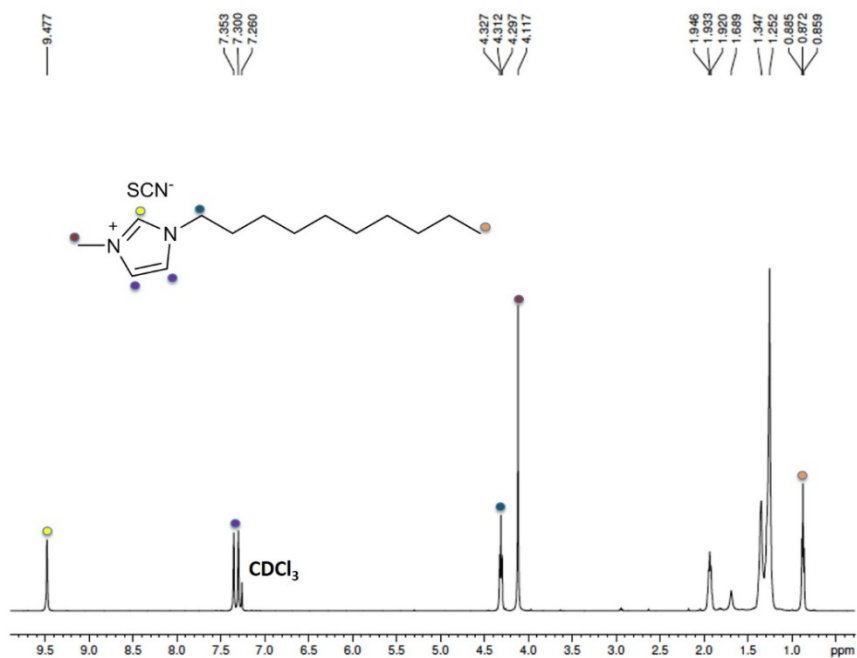
Figure S3B. ^{1}H NMR of dried 1-decyl-3-methylimidazolium thiocyanate, $C_{10}mim^+SCN^-$ in $CDCl_3$. (Sample contains a residual 4.9% (w/w) H_2O as determined by TGA.)
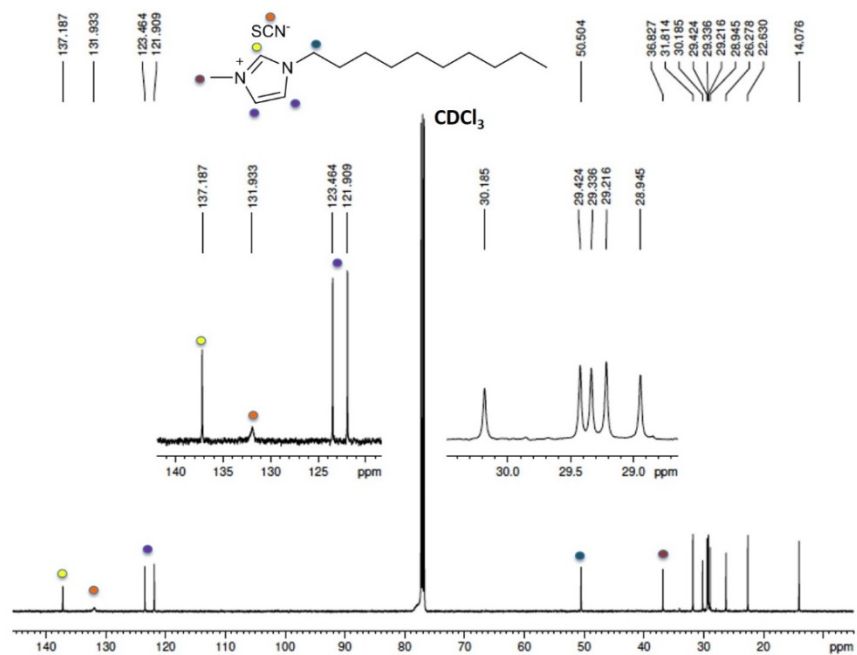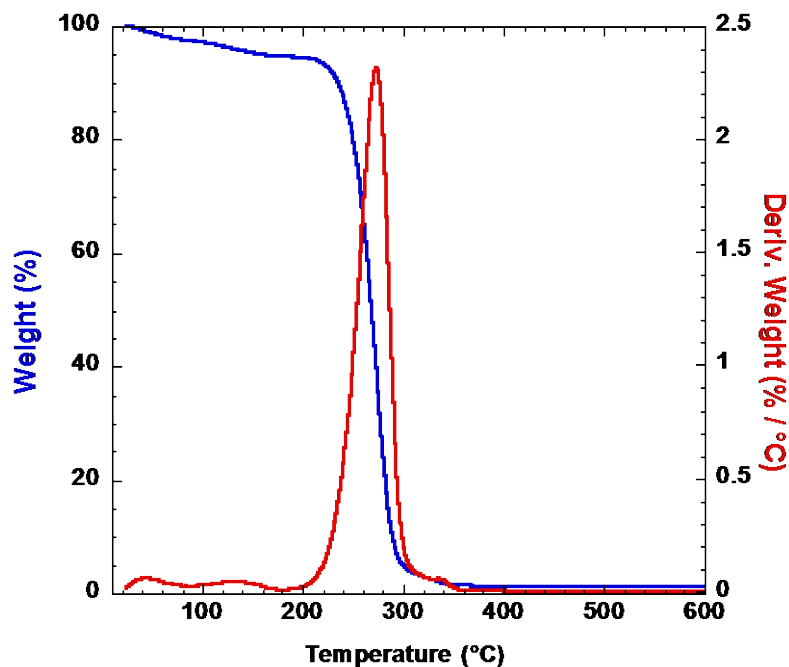

Figure S3C. ^{13}C NMR of dried 1-decyl-3-methylimidazolium thiocyanate, $C_{10}mim^+SCN^-$ in $CDCl_3$. (Sample contains a residual 4.9% (w/w) H_2O as determined by TGA).

Figure S4A. Fast scan (5 ^{o}C min^{-1}) N$_2$ atmosphere TGA collected on dried 1-decyl-3-methylimidazolium thiocyanate, C$_{10}$mim$^+$SCN$^-$. Thermogram shows 4.9% (w/w) H$_2$O remains in the sample.

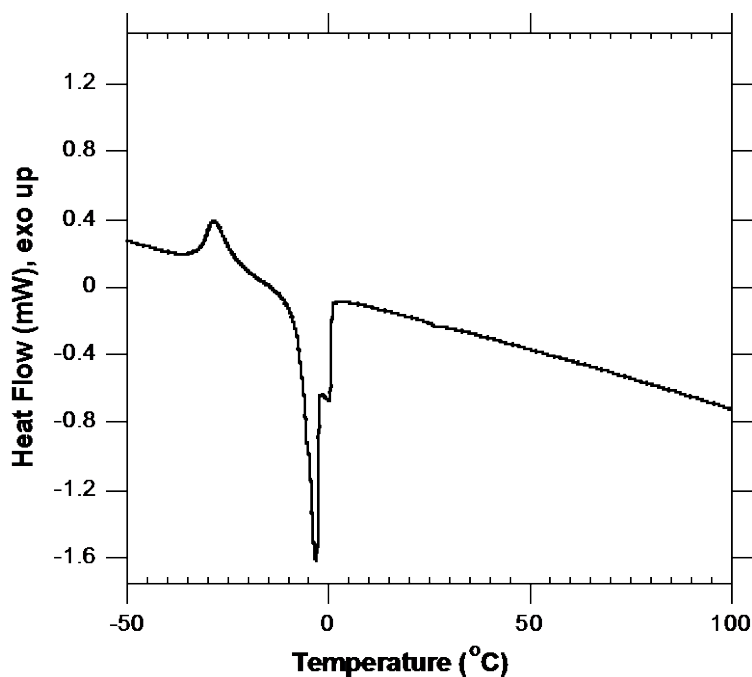---

Figure S4B. The second heating DSC profile (2 ^{o}C/min) collected on dried 1-decyl-3-methylimidazolium thiocyanate, C$_{10}$mim$^+$SCN$^-$. (Sample contained 4.9% (w/w) H$_2$O in the sample as determined by TGA.) Heating scan shows a T$_m$ = -6.74 ^{o}C.

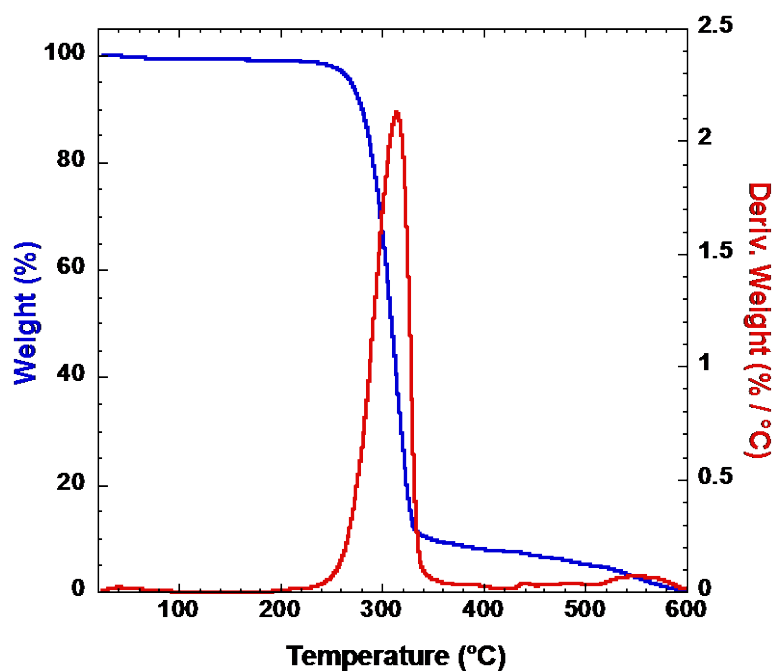**Figure S5A**. Fast scan (5 ^{o}C min^{-1}) N$_2$ atmosphere TGA collected on dried 1-decyl-3-methylimidazolium nitrate, C$_{10}$mim^{+}NO$_3$$^{-}$. Thermogram shows a residual 0.92% (w/w) H$_2$O remains in the sample.
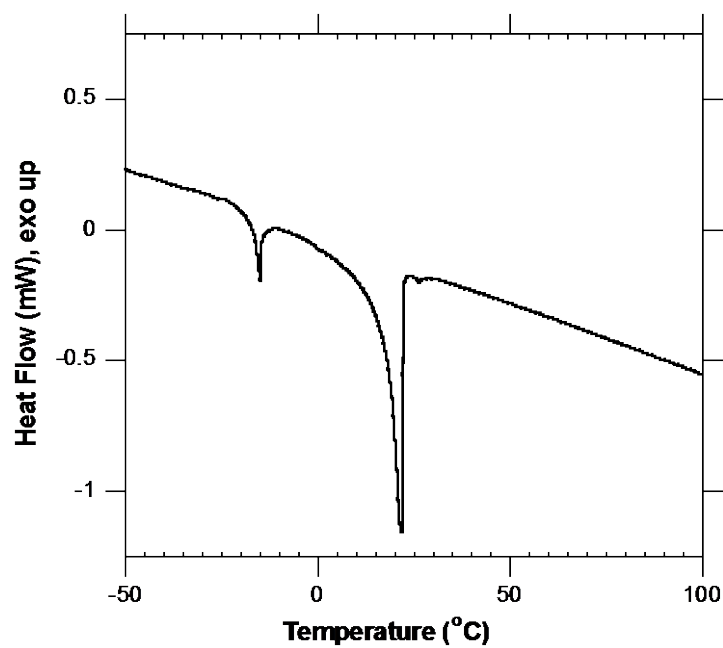

Figure S5B. The second heating DSC profile (2 ^{o}C/min) collected on dried (1-decyl-3-methylimidazolium nitrate, [C$_{10}$mim]$^{+}$[NO$_3$]$^{-}$. (Sample contained 0.92% (w/w) H$_2$O in the sample as determined by TGA.) Heating scan shows a T$_m$ = 17.8 ^{o}C.

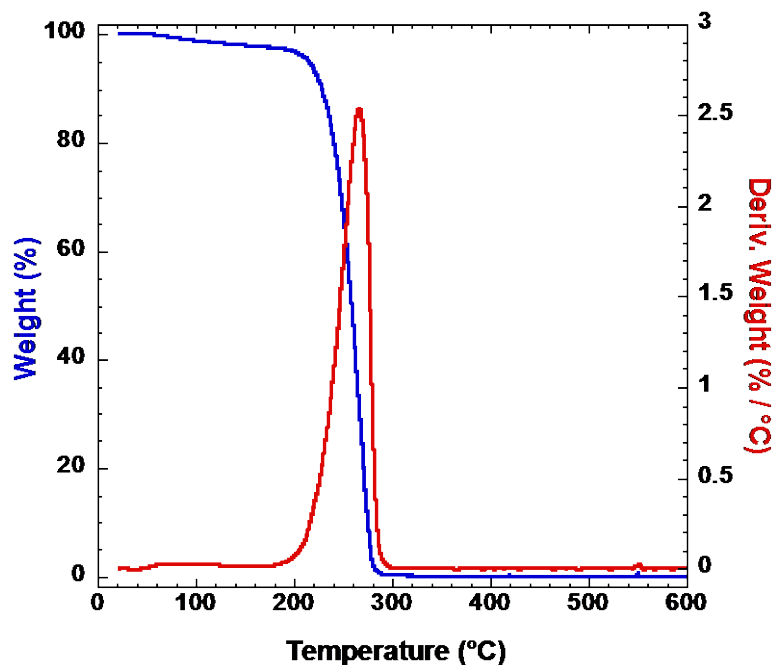**Figure S6A**. Fast scan (5 $^{\circ}$C min^{-1}) N$_2$ atmosphere TGA collected on dried 1-decyl-3-methylimidazolium chloride, C$_{10}$mim$^+$Cl$^-$. Thermogram shows 2% (w/w) H$_2$O remains in the sample.

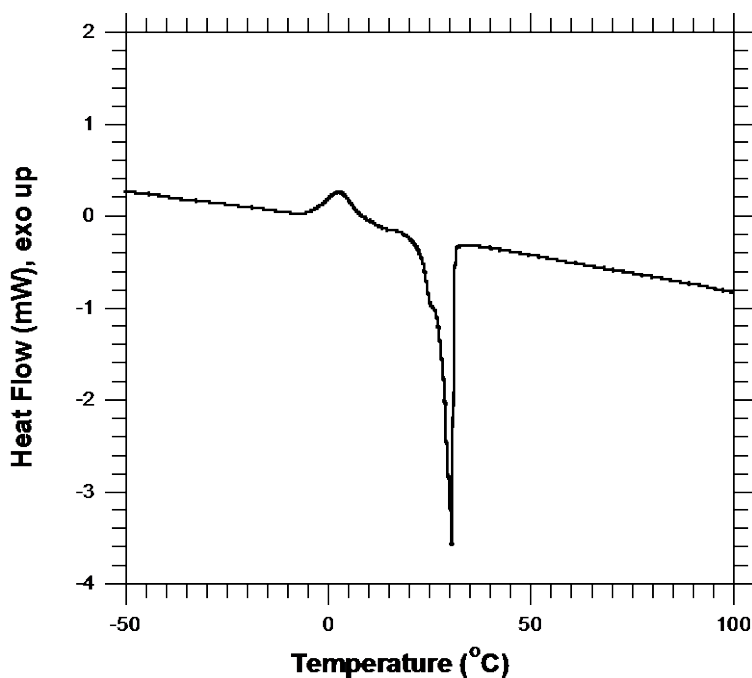**Figure S6B**. The second heating DSC profile (2 $^{\circ}$C/min) collected on dried (1-decyl-3-methylimidazolium chloride, C$_{10}$mim$^+$Cl$^-$. (Sample contained 2.0% (w/w) H$_2$O in the sample as determined by TGA.) Heating scan shows a T$_m$ = 27.5 $^{\circ}$ C.