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Supplementary Information

A new approach to very high lithium salt content quasi-solid state electrolytes for lithium metal batteries using plastic crystals

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Table S1. Transition temperatures (onset) and entropies for the $[C_2epyr][FSI]$/Li[FSI] salts, T_s = solid–solid transition, T_m = melting. Data for Li[FSI] obtained from ref [1]

	T_s (°C) ± 1	ΔS (J K^{-1} mol^{-1}) ± 10%	T_m(°C) ± 1	ΔS (J K^{-1} mol^{-1}) ± 10%
$[C_2epyr][FSI]$	-35	36	129	9
90mol% LiFSI in $[C_2epyr][FSI]$	-50	1	94	28
Li[FSI] [1]	-51	2	140	41

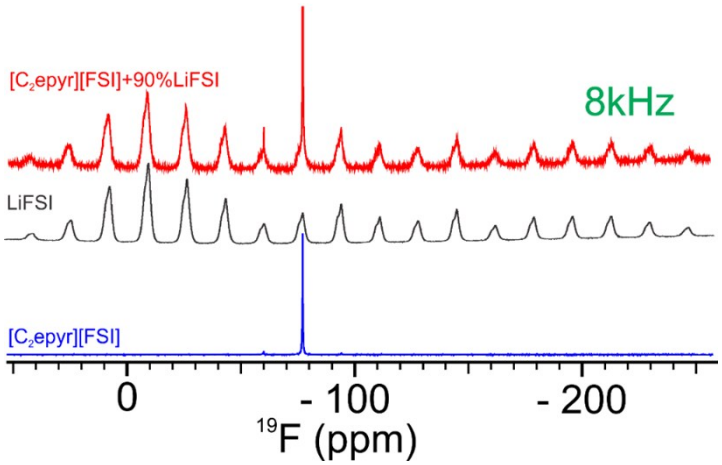

Figure S1. 8 kHz MAS ^{19}F NMR spectra of neat $[C_2epyr][FSI]$, neat Li [FSI] and 90 mol% Li[FSI] in $[C_2epyr][FSI]$.

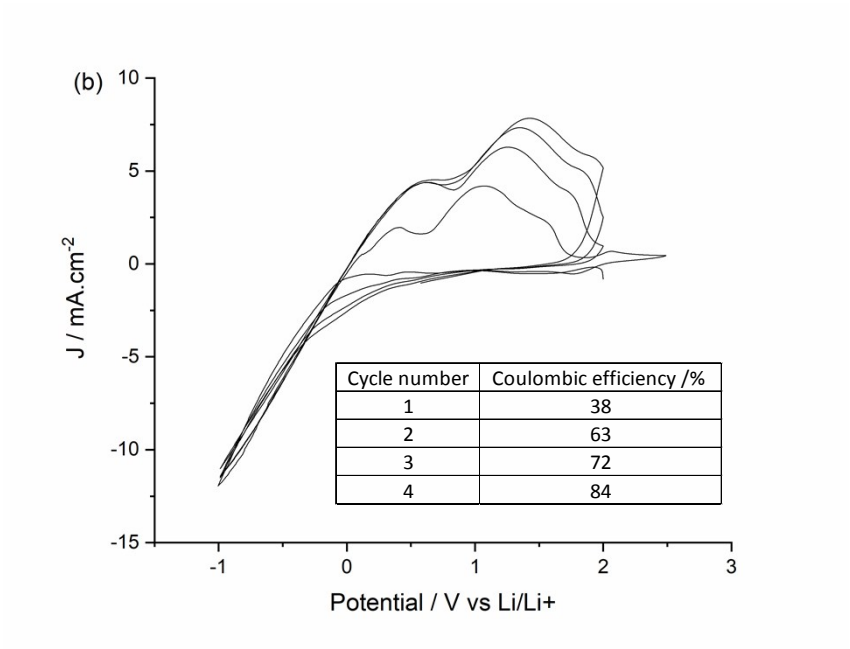

Figure S2. Cyclic voltammogram of the quasi-solid state 90 mol% Li[FSI] in [C_2epyr][FSI] electrolyte at 50℃. The arrows show the direction of peak progression from the 1st to the 4th scan, with platinum working electrode (2.0 mm^2 surface area), lithium strip as a quasi-reference electrode and a coiled lithium metal strip as counter electrode.

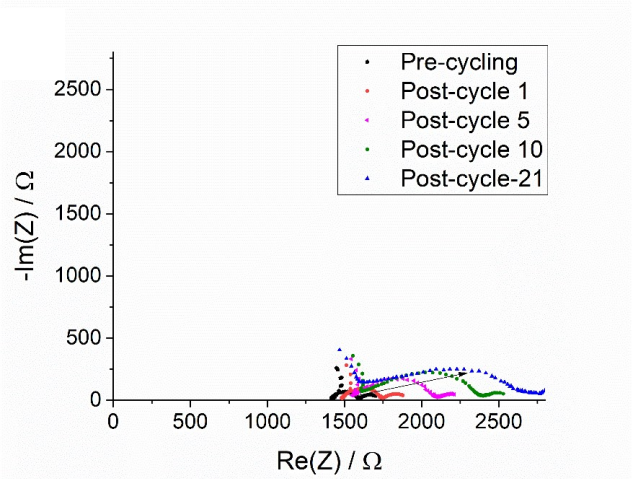

Figure S3. The Nyquist plots obtained after select plating cycles in a Li | Li symmetrical cell containing the quasi-solid 90 mol% Li[FSI] in [C_2epyr][FSI]. Cycling data shown in Figure 5 (a).

References.

[1] Y. Zhou, X. Wang, H. Zhu, M. Yoshizawa-Fujita, Y. Miyachi, M. Armand, M. Forsyth, G. W. Greene, J. M. Pringle, P. C. J. C. Howlett, *ChemSusChem* **2017**, 10, 3135.