

Electronic Supplementary Information

Lithium-Ion Battery Performance Enhanced by Combination of Si Thin Flake Anode and Binary Ionic Liquid System

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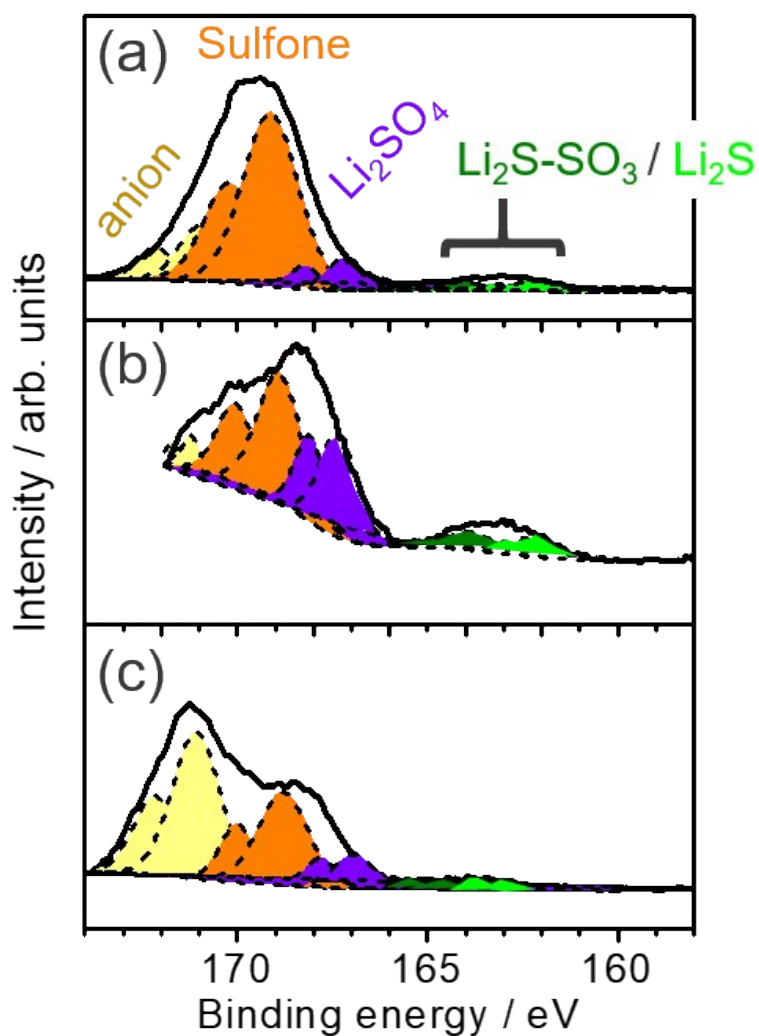


Fig. S1 XPS spectra for S-2p of the Si thin flake composite anodes after five charge-discharge cycles. The charge-discharge test was conducted by a CC mode at 0.5 C, and the cut-off voltages were -3.88 V and -2.40 V (vs. LiCoO_2). The electrolytes were (a) 83.3–16.7 mol% $[\text{C}_2\text{mim}][\text{FSA}]\text{-Li}[\text{TFSA}]$, (b) 83.3–16.7 mol% $[\text{C}_2\text{mim}][\text{FSA}]\text{-Li}[\text{FSA}]$, and (c) 50.0–50.0 mol% $[\text{C}_2\text{mim}][\text{FSA}]\text{-Li}[\text{FSA}]$.

Table S1 Densities of the IL electrolytes employed in this study at 298 K

IL electrolytes	$d / \text{g cm}^{-3}$
83.3–16.7 mol% [C ₂ mim][FSA]–Li[TFSA]	1.51
83.3–16.7 mol% [C ₂ mim][FSA]–Li[FSA]	1.49
50.0–50.0 mol% [C ₂ mim][FSA]–Li[FSA]	1.62

Table S2 Chemical species contained in the SEI films formed in different IL electrolytes and their composition ratios

IL electrolytes	Composition ratio / at%				
	LiF	Li ₃ N	Li ₂ S	Li ₂ SO ₄	-SO ₂ R
83.3–16.7 mol% [C ₂ mim][FSA]–Li[TFSA]	20.6	2.0	4.8	23.0	49.5
83.3–16.7 mol% [C ₂ mim][FSA]–Li[FSA]	63.3	0.9	5.8	18.8	11.3
50.0–50.0 mol% [C ₂ mim][FSA]–Li[FSA]	45.8	2.9	3.8	27.9	19.7

Table S3 Summary of Li-ion conductivities and shear modulus for potential SEI components

	Li ion conductivity $\sigma / \text{S cm}^{-1}$	Shear modulus G / GPa
LiF ^{S1,S2}	6×10^{-6} (323 K)	55
Li ₃ N ^{S3,S4}	1×10^{-3} (300 K)	64
Li ₂ SO ₄ ^{S5,S6}	5×10^{-8} (298 K)	20
Li ₂ S ^{S7-S9}	$> 10^{-11}$ (298 K)	32~35
LiPON ^{S10}	2×10^{-6} (298 K)	31

Captions for Movie S1 and S2

Movie S1 Video clip of a binder free Si thin flake anode in the *operando* SEM observation cell with a 83.3–16.7 mol% [C₂mim][FSA]–Li[TFSA] IL electrolyte during the fourth charge process. The charge process was conducted in a CC/CV mode with cut-off voltages ranging between –3.88 V and –2.40 V (vs. LiCoO₂). The CC rates for charge and discharge were 1/2 C. The cut-off current density was 1/20 C. The video clip plays at 700 × speed.

Movie S2 Video clip of a binder free Si thin flake anode in the *operando* SEM observation cell with a 83.3–16.7 mol% [C₂mim][FSA]–Li[FSA] IL electrolyte during the fourth charge process. The charge process was conducted in a CC/CV mode with cut-off voltages ranging between –3.88 V and –2.40 V (vs. LiCoO₂). The CC rates for charge and discharge were 1/2 C. The cut-off current density was 1/20 C. The video clip plays at 700 × speed.

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