

Supplementary Materials For

Understanding steric hindrance effect of solvent molecule in localized high-concentration electrolyte for lithium metal batteries

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Supplementary Figures

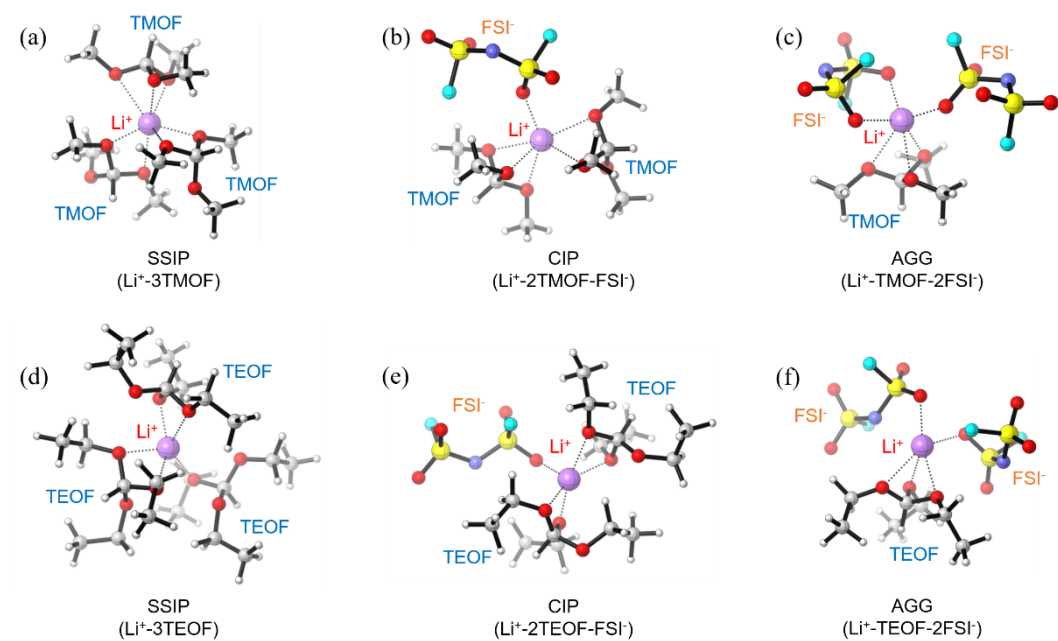


Fig. S1 DFT optimized solvation structures in the (a-c) LiFSI-1.2TMOF-3TTE and (d-f) LiFSI-1.2TEOF-3TTE electrolytes.

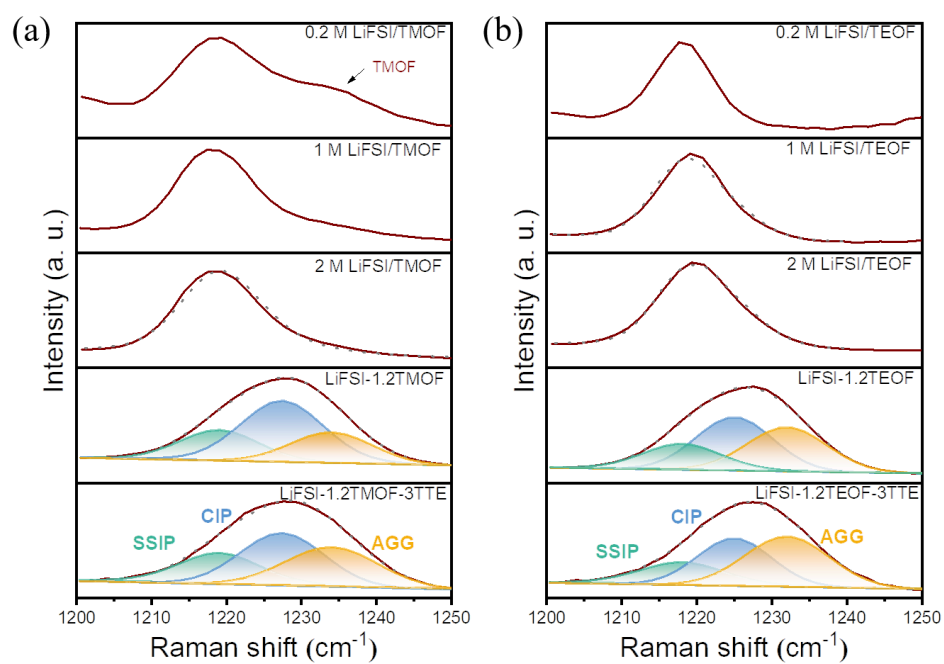


Fig. S2 Raman spectra of the (a) TMOF-based and (b) TEOF-based electrolytes.

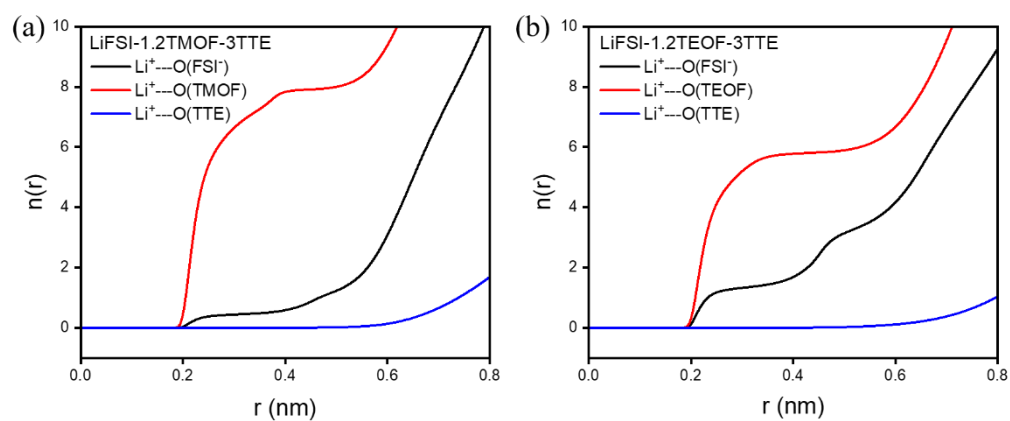


Fig. S3 Integrated coordination number calculated from MD simulations of the **(a)** TMOF-based and **(b)** TEOF-based electrolytes.

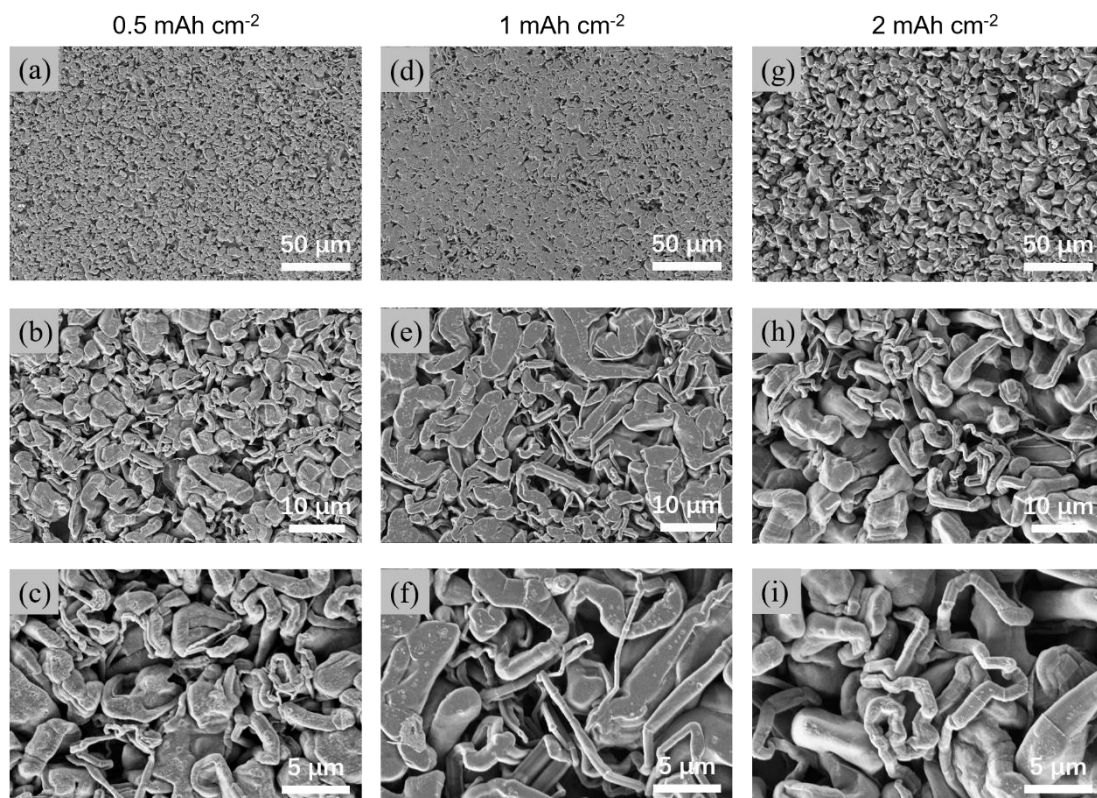


Fig. S4 SEM images of the deposits in LiFSI-1.2TMOF-3TTE electrolyte with a deposition capacity of (a-c) 0.5 mAh cm⁻², (d-f) 1 mAh cm⁻² and (g-i) 2 mAh cm⁻² at a current density of 0.5 mA cm⁻².

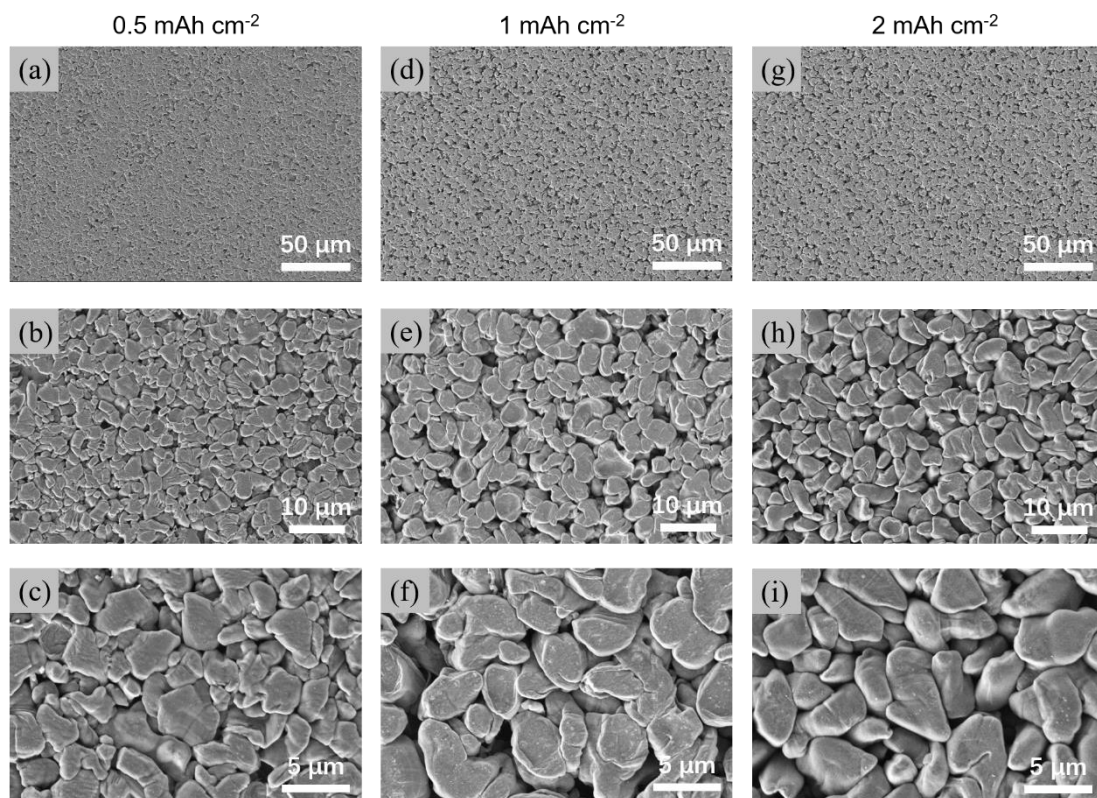


Fig. S5 SEM images of the deposits in LiFSI-1.2TMOF-3TTE electrolyte with a deposition capacity of (a-c) 0.5 mAh cm⁻², (d-f) 1 mAh cm⁻² and (g-i) 2 mAh cm⁻² at a current density of 0.5 mA cm⁻².

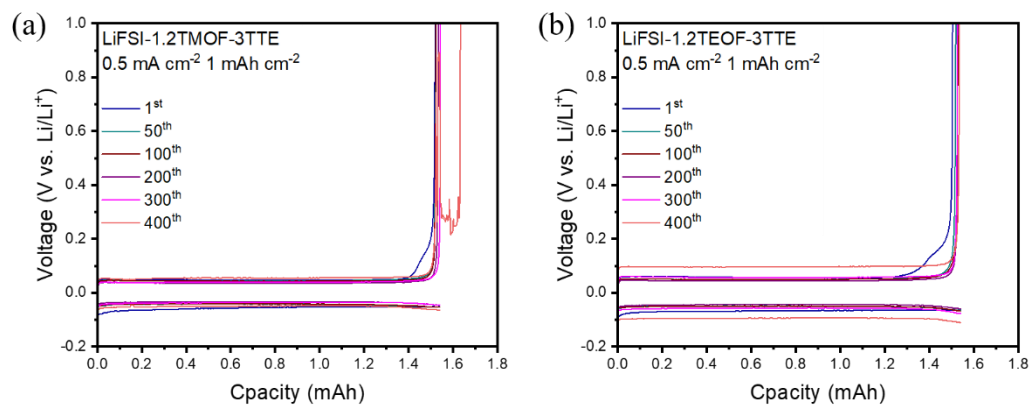


Fig. S6 Voltage profiles of Li||Cu cells cycled at 0.5 mA cm⁻² and 1 mAh cm⁻² in the (a) LiFSI-1.2TMOF-3TTE and (b) LiFSI-1.2TEOF-3TTE electrolyte.

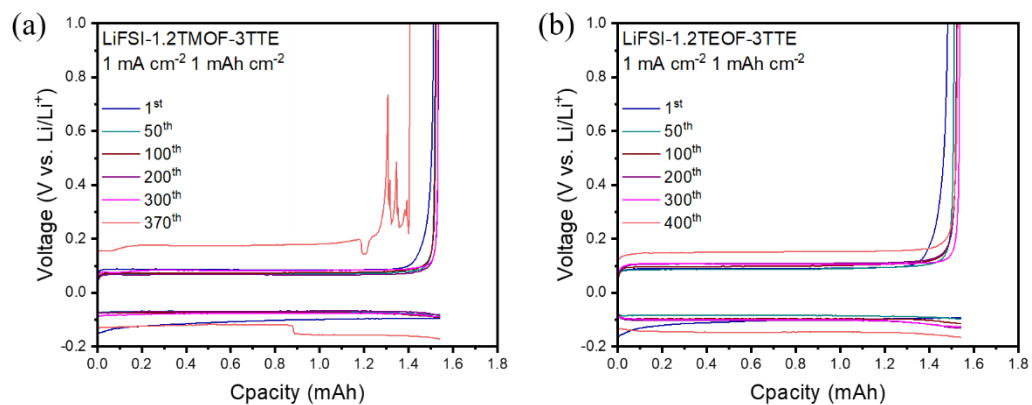


Fig. S7 Voltage profiles of Li||Cu cells cycled at 1 mA cm^{-2} and 1 mAh cm^{-2} in the (a) LiFSI-1.2TMOF-3TTE and (b) LiFSI-1.2TEOF-3TTE electrolyte.

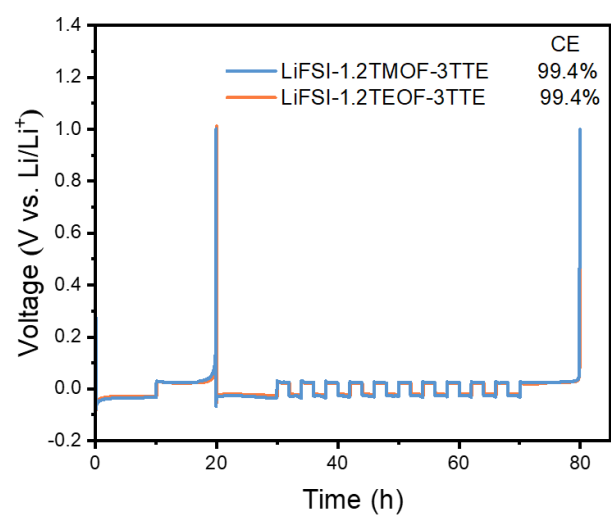


Fig. S8 Coulombic efficiency of Li||Cu cells tested using the Aurbach method.

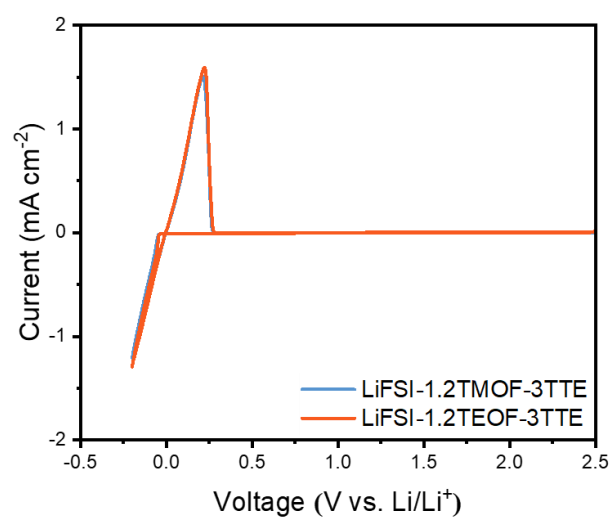


Fig. S9 Cyclic voltammetry (CV) results of Li||Cu cells cycled in two electrolytes.

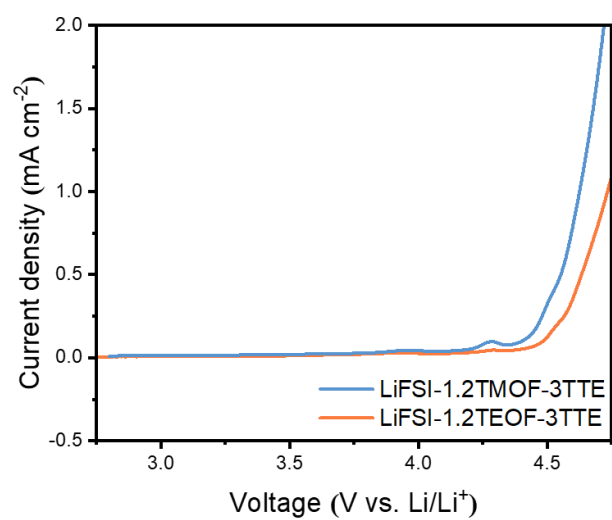


Fig. S10 Linear sweep voltammetry (LSV) measurements of Li||carbon paper cells cycled in two electrolytes.

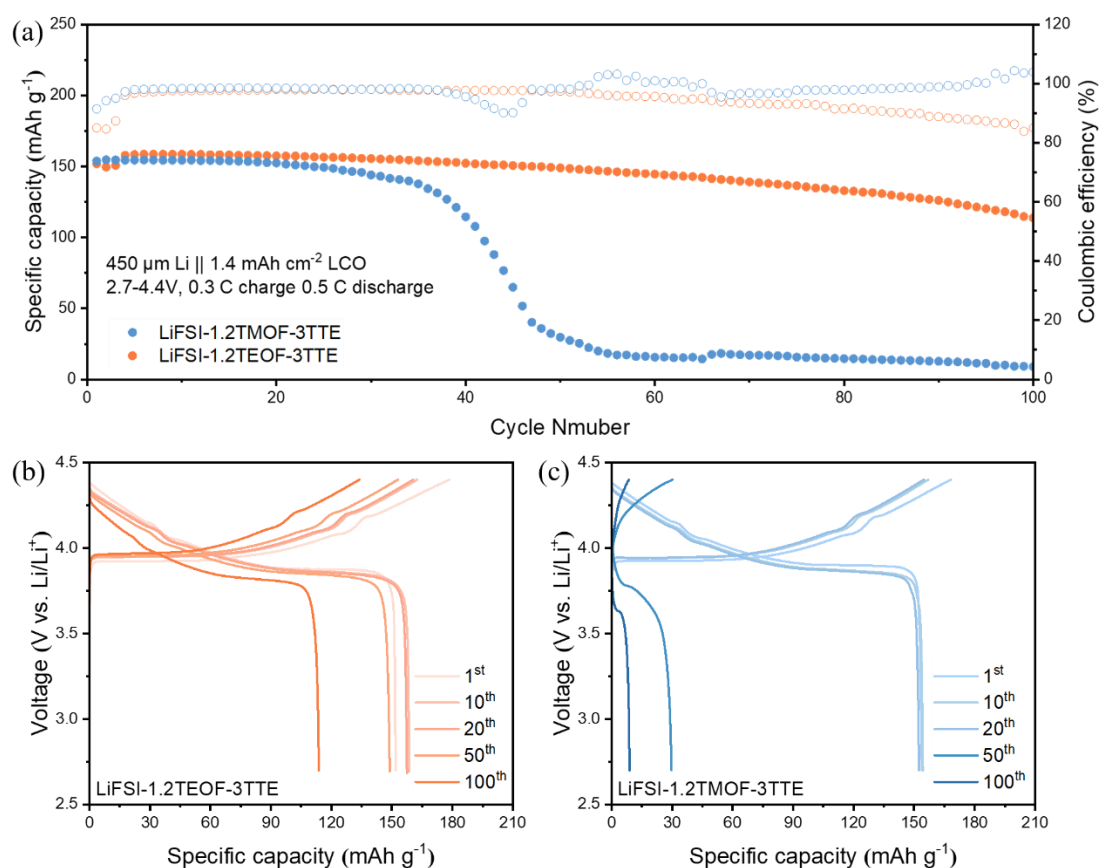


Fig. S11 (a) Long-term cycling performances of Li||LCO cells in the two electrolytes. The corresponding capacity-voltage profiles of the cell cycled in the (b) LiFSI-1.2TEOF-3TTE and (c) LiFSI-1.2TMOF-3TTE electrolyte.

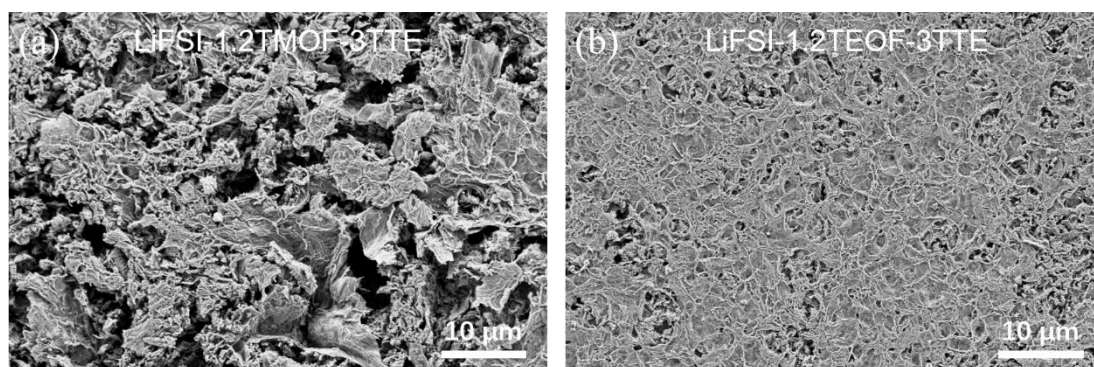


Fig. S12 The morphology of Li metal anode cycled in Li||NMC811 cells for 50 cycles using the (a) LiFSI-1.2TMOF-3TTE and (b) LiFSI-1.2TEOF-3TTE electrolyte.

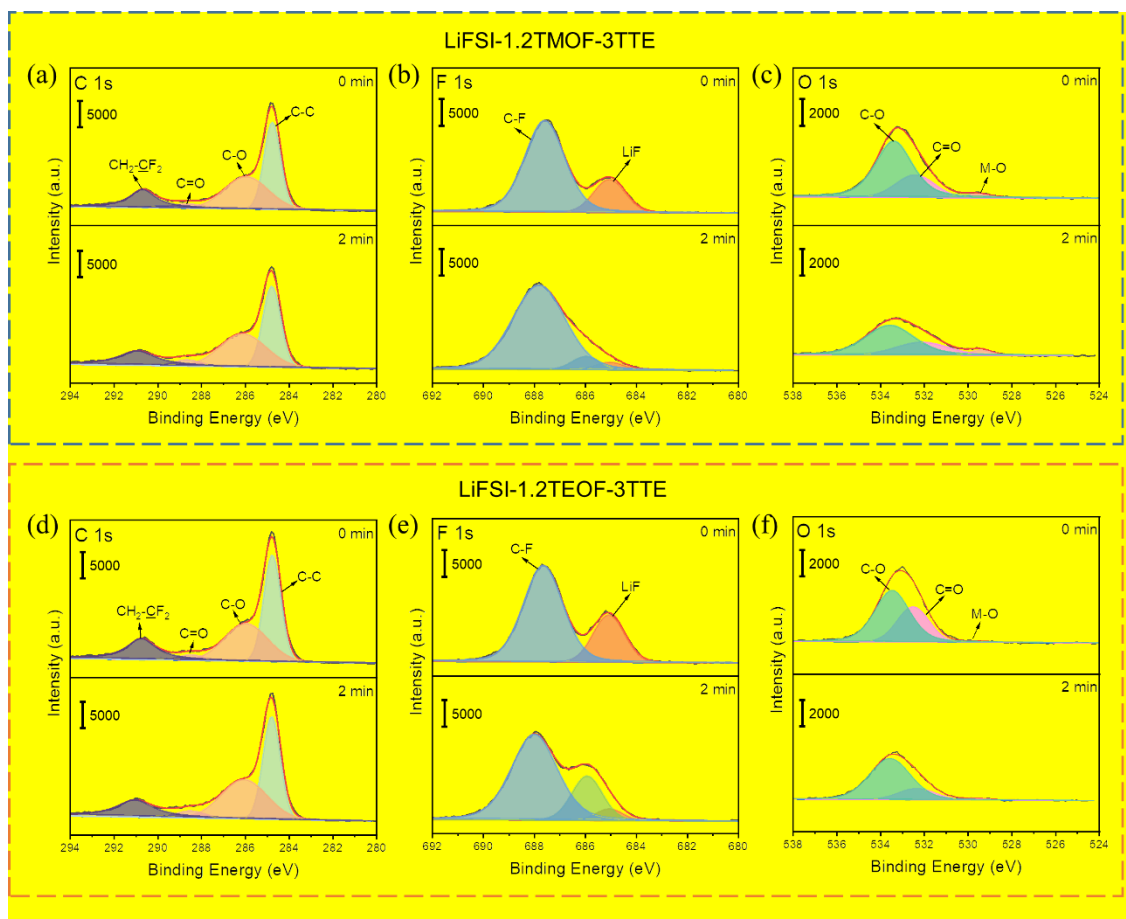


Fig. S13 XPS analysis of the NMC811 cathodes cycled in Li||NMC811 cells after 50 cycles using the (a-c) LiFSI-1.2TMOF-3TTE and (d-f) LiFSI-1.2TEOF-3TTE electrolyte. (a,d) C 1s, (b,e) F 1s and (c,f) O 1s XPS spectra are shown.

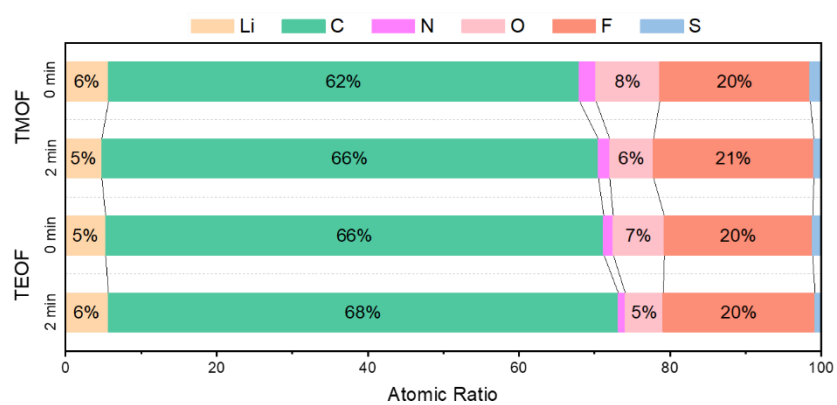


Fig. S14 Elemental ratios of the CEI formed on the cathode cycled in Li||NMC811 cells for 50 cycles.