## Supporting Information

Engineering the Solid Electrolyte Interphase for Enhancing High-Rate Cycling and Temperature Adaptability of Lithium-Ion Batteries

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**Keywords:** Lithium-ion Battery; Electrolyte Additive; Lithium Nonafluoro-1-Butanesulfonate; Electrode/Electrolyte Interface.

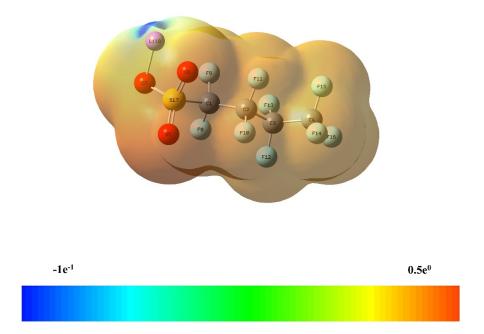


Figure S1. Electron cloud density distribution of NFSALi.

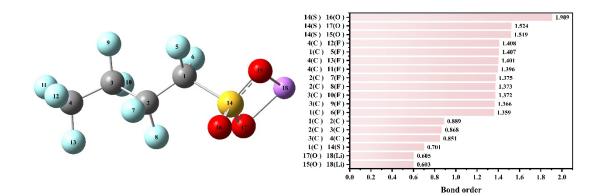


Figure S2. Bond order of NFSALi

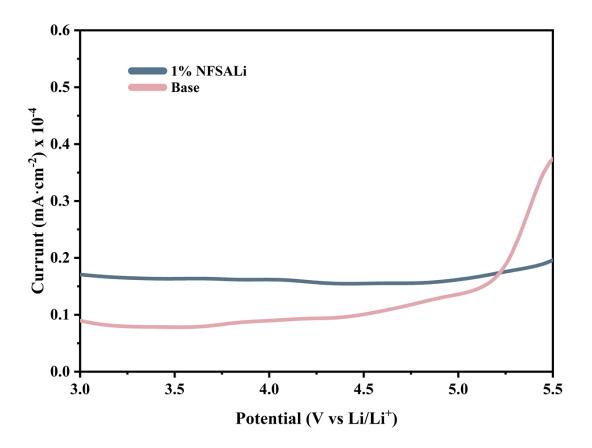


Figure S3. Oxidation potential by LSV with steel foil as work electrode, Li foil as counter- and reference electrodes at a scanning rate of  $1 \text{ mV s}^{-1}$ .

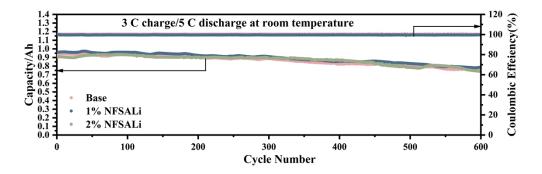
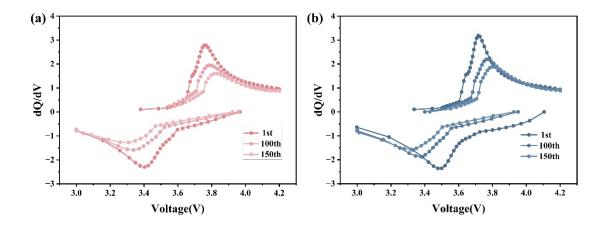
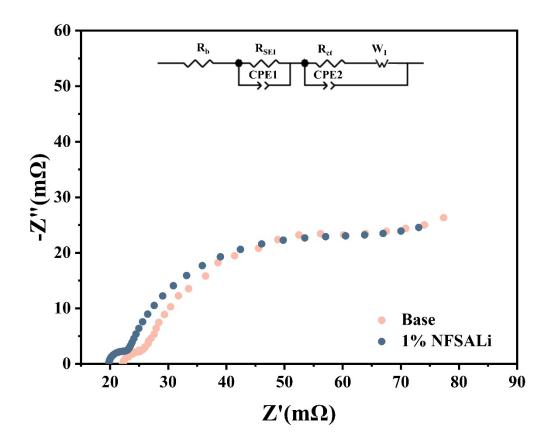


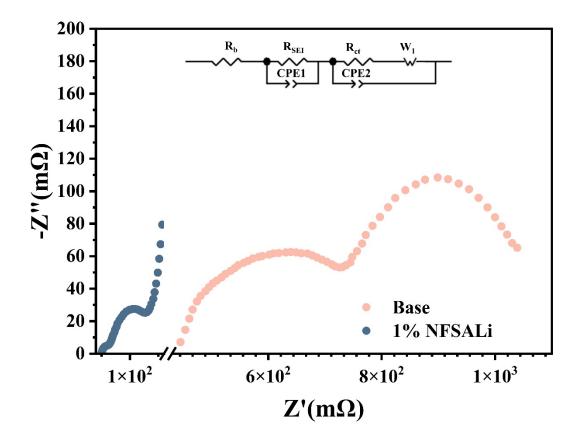
Figure S4. Cycling performance under 3 C charge/5 C discharge rate at room temperature.



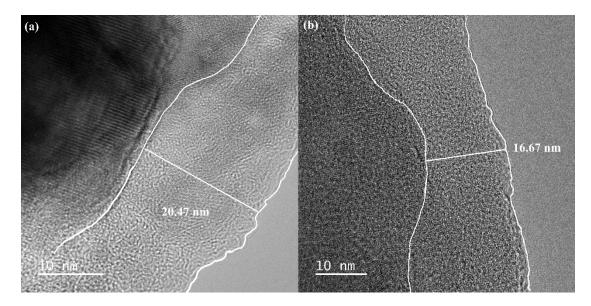
**Figure S5.** The dQ/dV curves of various electrolytes with different cycles at 55 °C, (a) base electrolyte, (b) 1% NFSALi electrolyte.



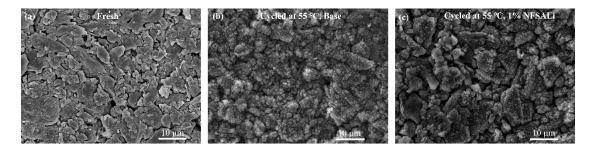
**Figure S6**. The electrochemical impedance spectroscopy (EIS) of each electrolyte before cycling.



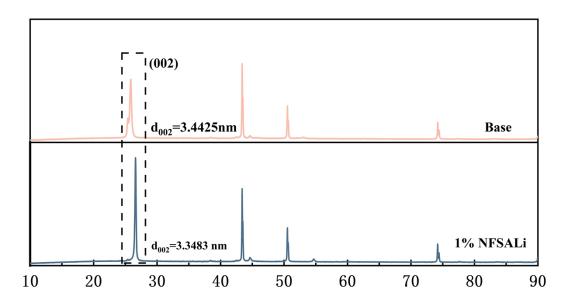
**Figure S7**. The electrochemical impedance spectroscopy (EIS) of each electrolyte after cycling at 55 °C.



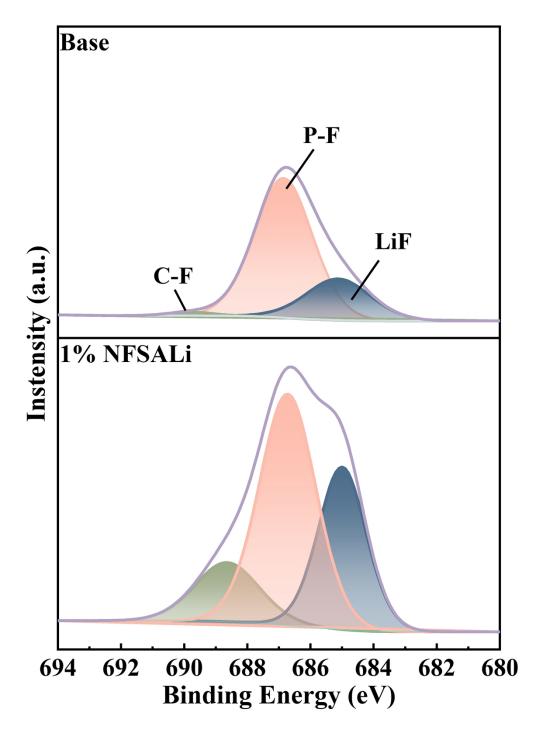
**Figure S8.** Transmission Electron Microscope (TEM) image of the NCM523 electrodes: (a) base electrolyte after 200 cycles at 55 °C, (b) 1% NFSALi electrolyte after 200 cycles at 55 °C.



**Figure S9.** Surficial SEM images of graphite electrodes: (a) fresh, (b) after 200 cycles at 55 °C with base electrolyte, (c) after 200 cycles at 55 °C with 1% NFSALi electrolyte.



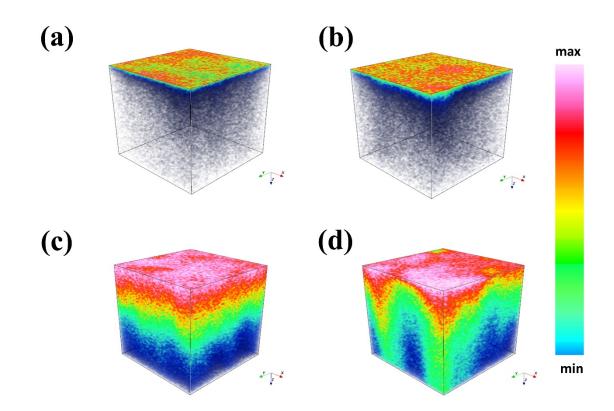
**Figure S10.** XRD patterns of graphite electrodes after cycling at 55 °C in base electrolyte and NFSALi containing electrolyte.



**Figure S11** The X-ray photoelectron spectroscopy (XPS) analysis of the NCM523 electrodes: F 1s spectra after cycling.

Regions	Component	Base	1% NFSALi
F 1s	P-F	74.17	31.63
	C-F	1.47	15.41
	LiF	24.36	31.63

 Table S1 Atomic concentration (%) of NCM523 electrodes after 200 cycles.



**Figure S12.** 3D render of  $PO_2^{-}$ :(a) base electrolyte, (b) 1% NFSALi electrolyte, (c) base electrolyte after 200 cycles at 55 °C, (d) 1% NFSALi electrolyte after 200 cycles at 55 °C.