Supporting Information for

Insights Into the Interfacial Degradation of High-Voltage All-Solid-State Lithium Batteries

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Supplementary Figures and Tables

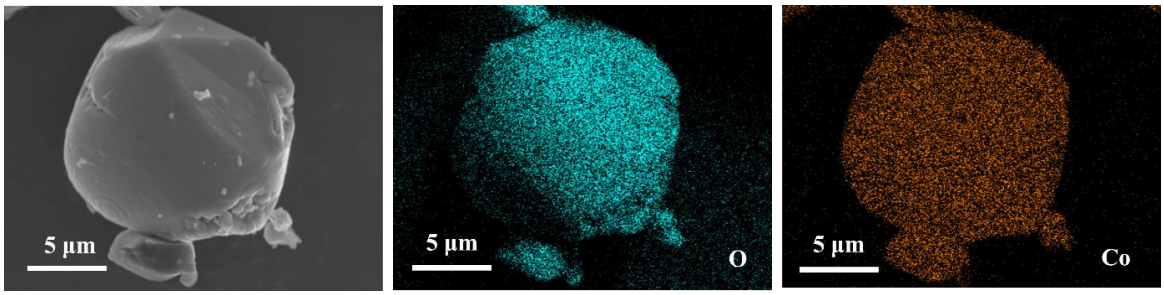

Fig. S1 EDS elemental maps of Co and O in a LCO particle

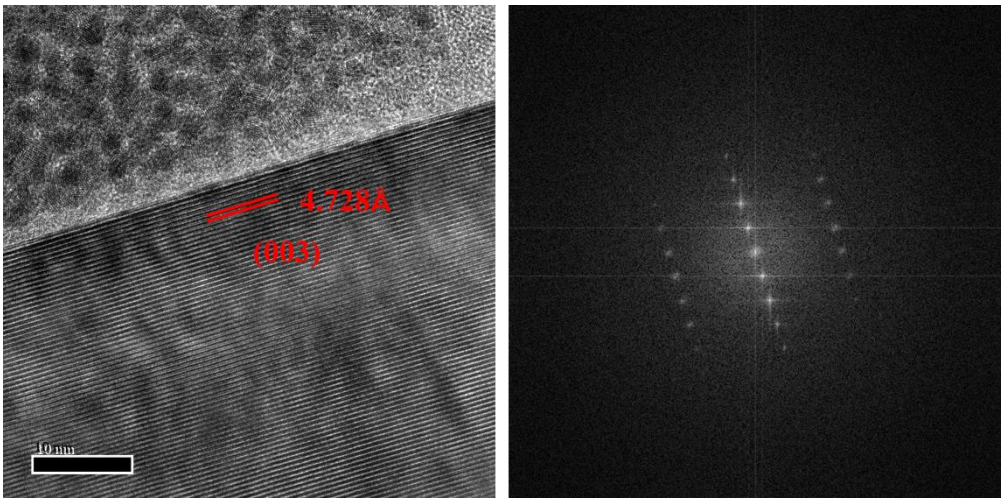

Fig. S2 TEM image and FFT pattern of LCO particle

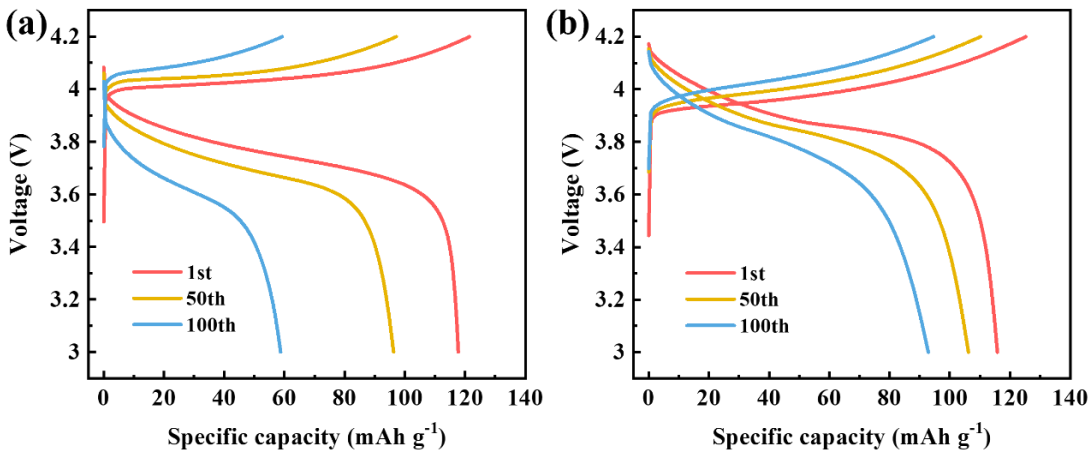

Fig. S3 Charge/discharge profiles of **a** LCO/PEO-LiTFSI/Li, **b** LAF@LCO/PEO-LiTFSI/Li cell at 0.5 C

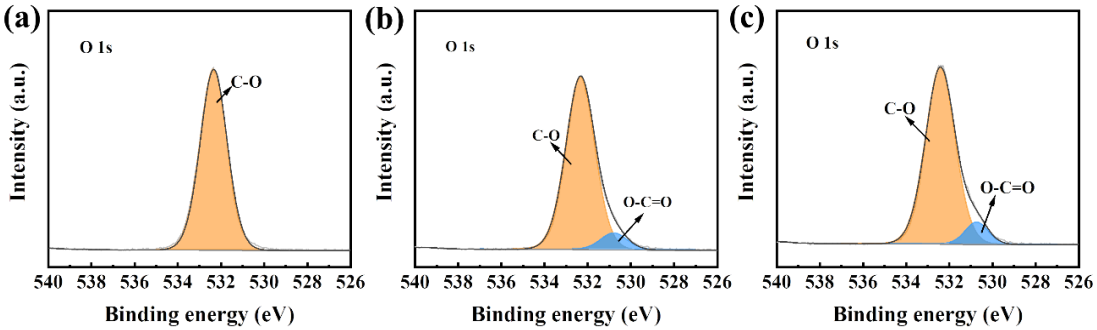

Fig. S4 O 1s from **a** pristine PEO-based SPE, PEO-based SPE surface after cycling with **b** LCO electrode and **c** LAF@LCO electrode. Cycled samples were obtained from the ASSLBs after 50 cycles in voltage range of 3.0-4.2 V

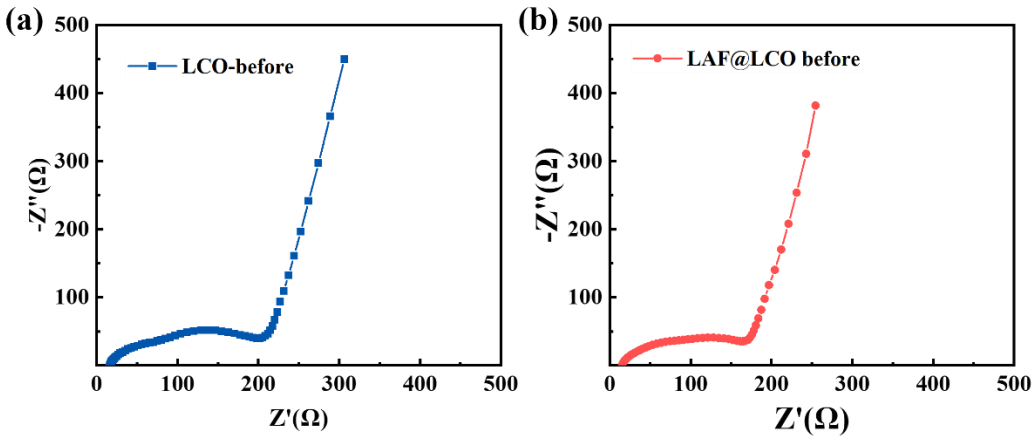

Fig. S5 EIS spectra of **a** LCO/PEO-LiTFSI/Li celland **b** LAF@LCO/PEO-LiTFSI/Li cell before cycling

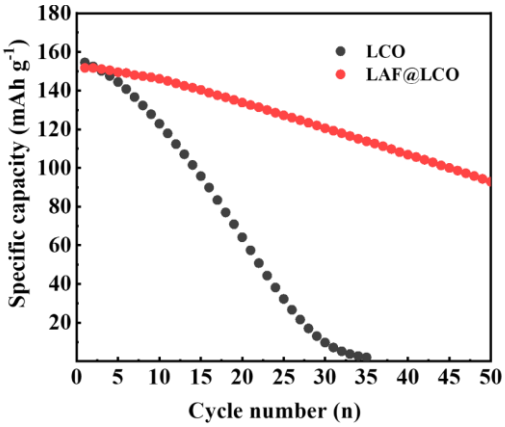

Fig. S6 The cycling performance of LCO/PEO-LiTFSI/Li and LAF@LCO/PEO-LiTFSI/Li cell, respectively. The cells were charged at a constant current of 1 C to 4.5 V, and followed by discharging to 3.0 V at 0.2 C at 60 °C

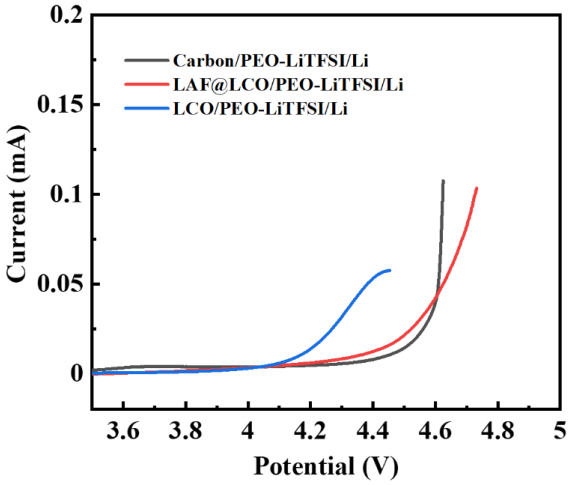

Fig. S7 Comparison of the linear sweep voltammogram of the carbon/PEO-LiTFSI/Li, LCO/PEO-LiTFSI/Li and LAF@LCO/PEO-LiTFSI/Li cell. The scan rate is 0.1 mV s^{-1}

Table S1 Performance of LCO cathode with different coating materials in PEO-based solid-state batteries

Coating material	wt% of Coating material	Cut off voltage	Capacity retention	Refs.
Li_3AlF_6	2%	4.5 V	85%/20 cycles 68%/50 cycles	Nano-Micro Lett. 2022, 10912
lithium tantalate		4.5 V	85%/20 cycles	J. Mater. Chem. A, 2020, 8, 2769–2776
LAGP		4.5 V	80%/7cycles	ACS Energy Lett. 2020, 5, 826−832
Li_3PO_4	5%	4.6 V	50%/9 cycles	Chem. Mater., 17, 8, 2005
PECA		4.45 V	40%/100 cycles	J. Electrochem. Soc., 2017, 164 (14), A3454-A3461
Al_2O_3	3%	4.4 V	65%/100 cycles	Chem. Mater., 2005, 17, 23
Li_3PO_4	5%	4.3 V	76%/20 cycles	Chem. Mater., 2005, 17, 8,
LATP		4.2 V	93.2%/50cycles (Coulomb efficiency<85%)	J. Power Sources 2018, 388 65–70
LATP		4.2 V	88%/50cycles	Adv. Funct. Mater. 2020, 30, 1909392
Li_3InCl_6	15%	4.2 V	68.4%/200cycles	Nano Energy 2020, 76 105015
LiDFOB-PVC		4.2 V	71.5%/500cycles	Energy Storage Mater. 2020, 32 191–198
Li_3AlF_6	2%	4.2 V	92.2%/50 cycles 80.2%/100 cycles	Nano-Micro Lett. 2022, 10912