Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.
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Electronic Supplementary Information

Ferroelectric $BiFeO_3$ modified PVDF-based electrolytes for high-performance lithium metal batteries

Yanmei Wu, Hong Zhang, Yilin Xu, Zhiruo Tang, Zhicheng Li*

School of Materials Science and Engineering, Central South University, Changsha 410083, China

Corresponding author: zhchli@csu.edu.cn

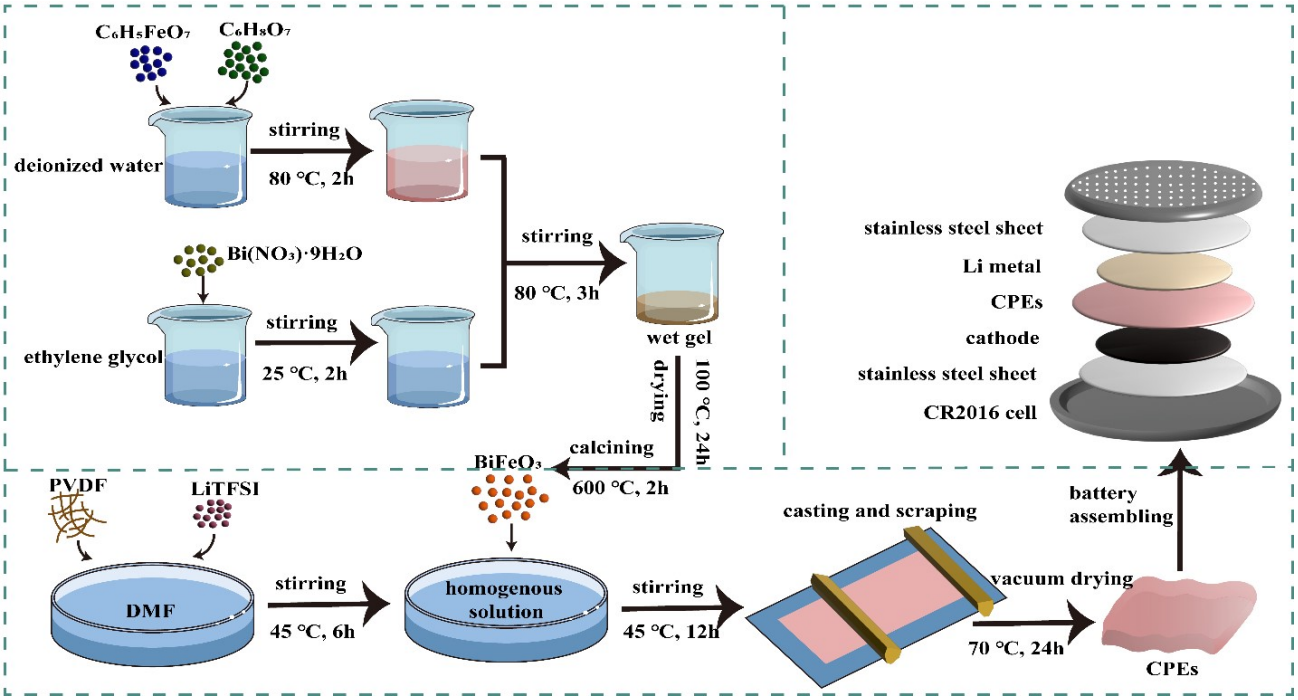
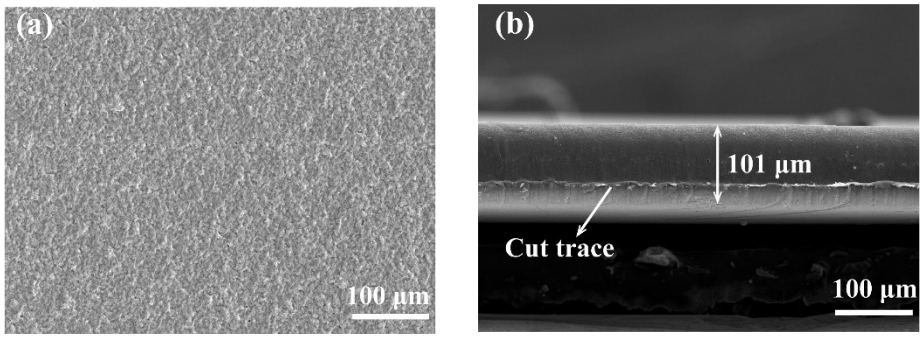


Fig. S1 Schematic diagram of the preparation process



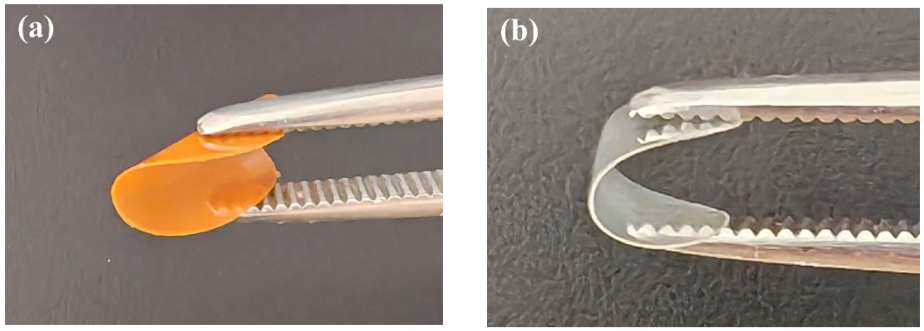


Fig. S2 SEM observation of PVDF film, (a) surface morphology, (b) cross-sectional view

Fig. S3 Optional images of PVDF-based films, (a) PVDF-5BFO, (b) PVDF

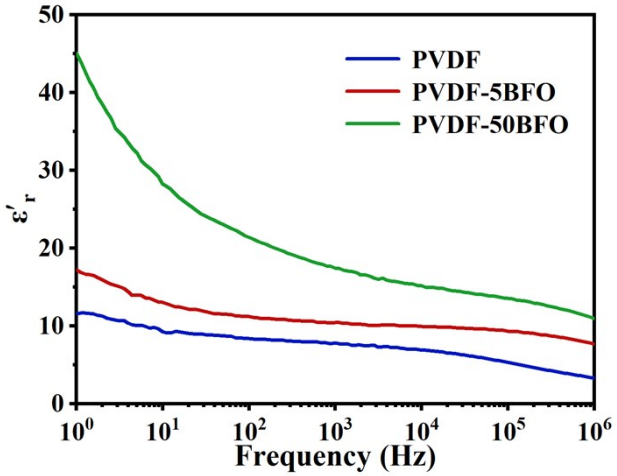


Fig. S4 Plots for frequency-dependent relative permittivity of PVDF-based films at 25 ℃

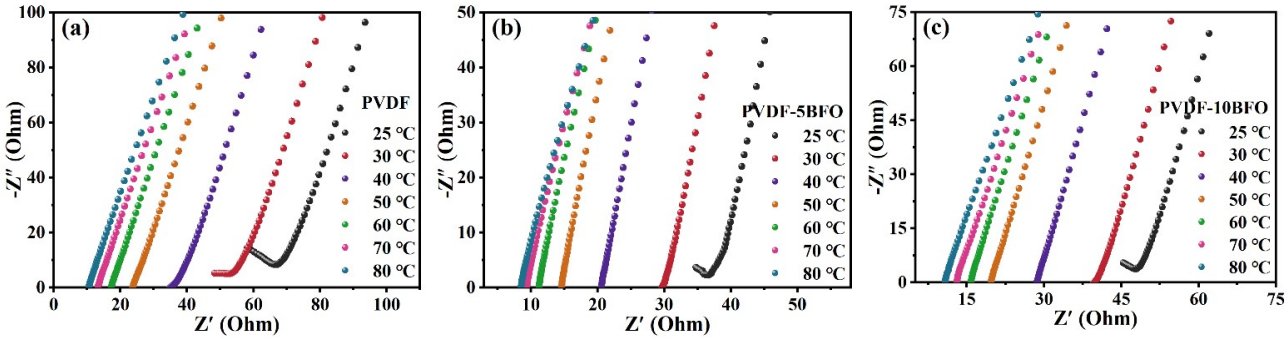
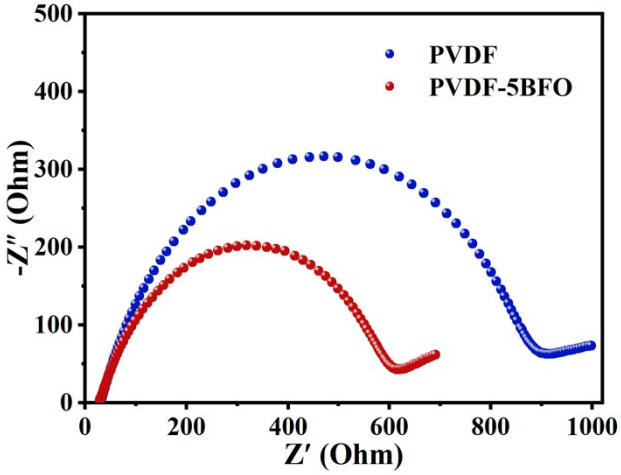


Fig. S5 Electrochemical impedance spectra of PVDF-based films at various temperatures, (a) PVDF, (b) PVDF-5BFO,



(c) PVDF-10BFO

Fig. S6 Electrochemical impedance spectra of Li|CPE|Li batteries after 100 cycles

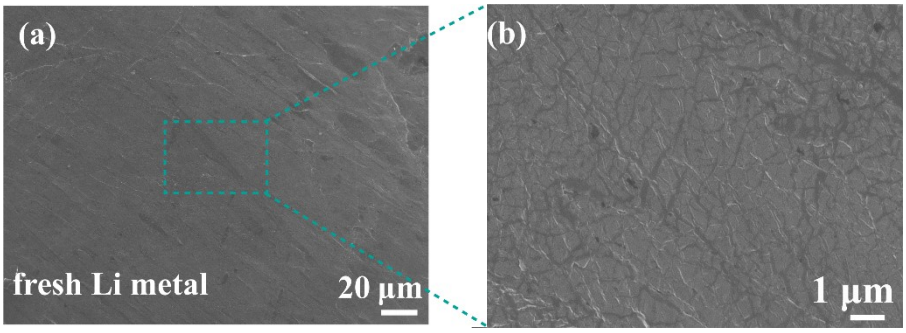


Fig. S7 SEM surface observation of fresh Li metal before cycling, (a) Li metal, (b) corresponding enlarged region

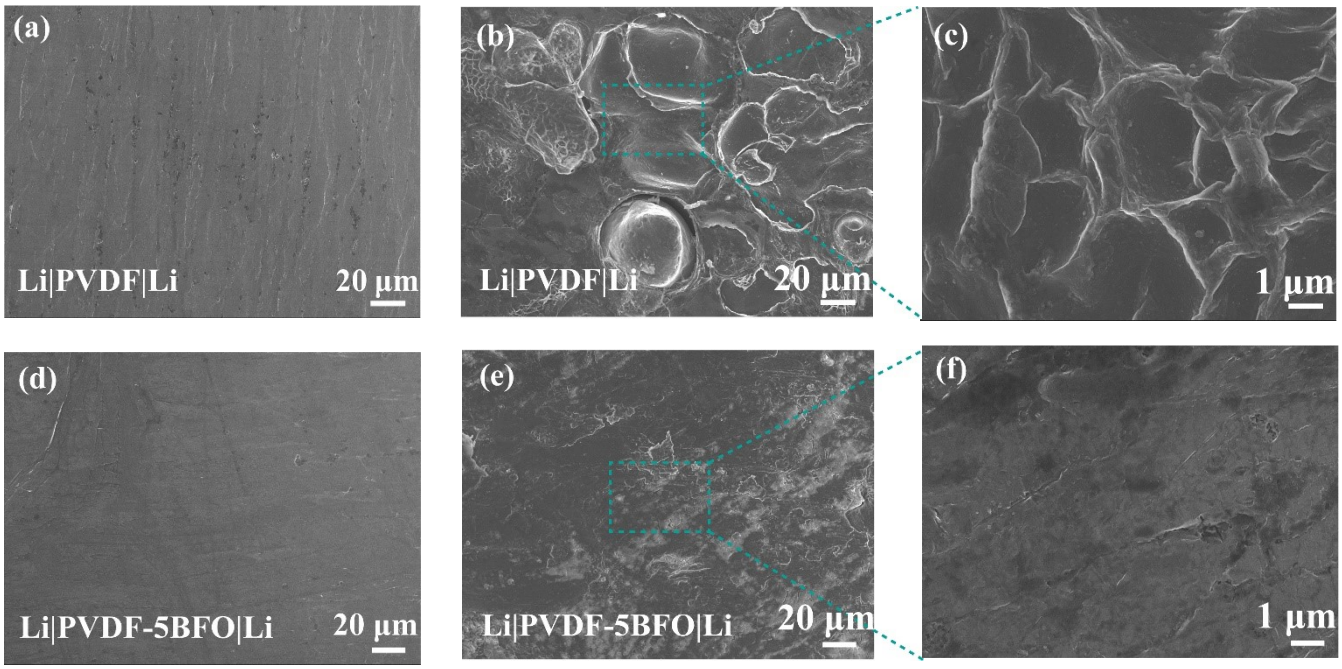


Fig. S8 SEM observation of the surfaces of Li-metals in symmetric Li|CPE|Li batteries after galvanostatic cycling, with two kinds of electrolytes, (a) PVDF as electrolyte and after 100 h, (b) PVDF as electrolyte after battery failure, (c) the corresponding enlarged Li metal region of Li|PVDF|Li, (d) PVDF-5BFO as electrolyte after 100 h, (e) PVDF-5BFO as electrolyte after 2500 h, (f) the corresponding enlarged Li metal region of Li|PVDF-5BFO|Li

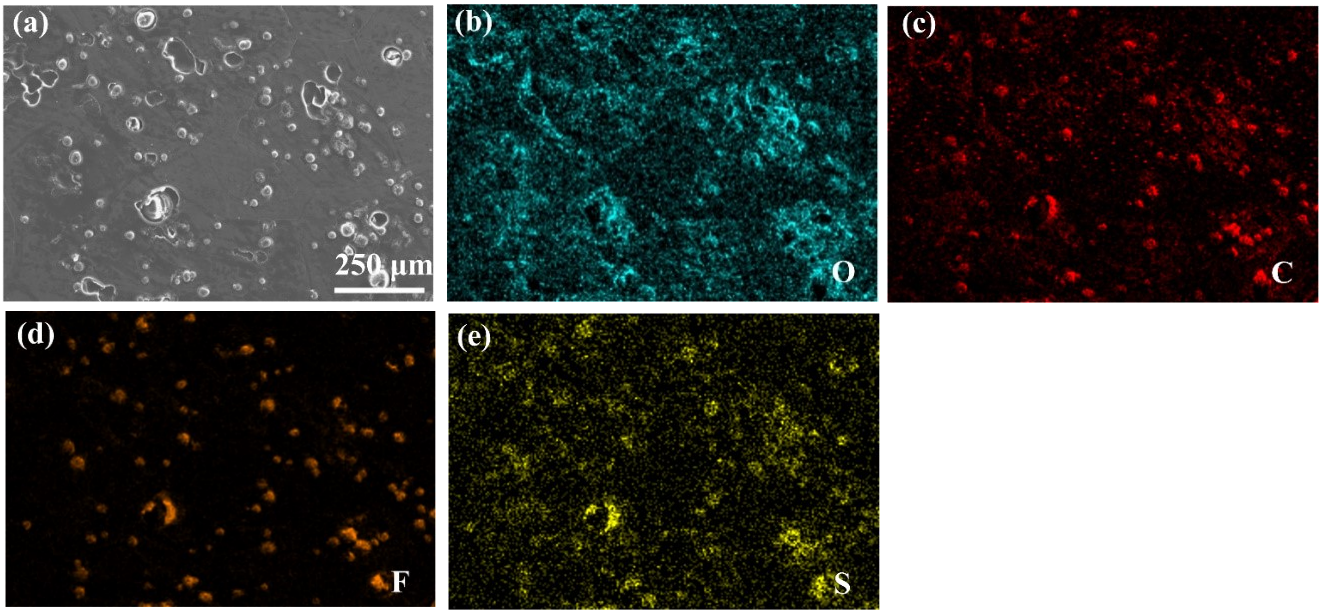


Fig. S9 Surface SEM image and corresponding EDS mapping of elemental distribution of cycled Li surface in Li|PVDF|Li battery, (a) surface SEM image, (b) O element, (c) C element, (d) F element, (e) S element

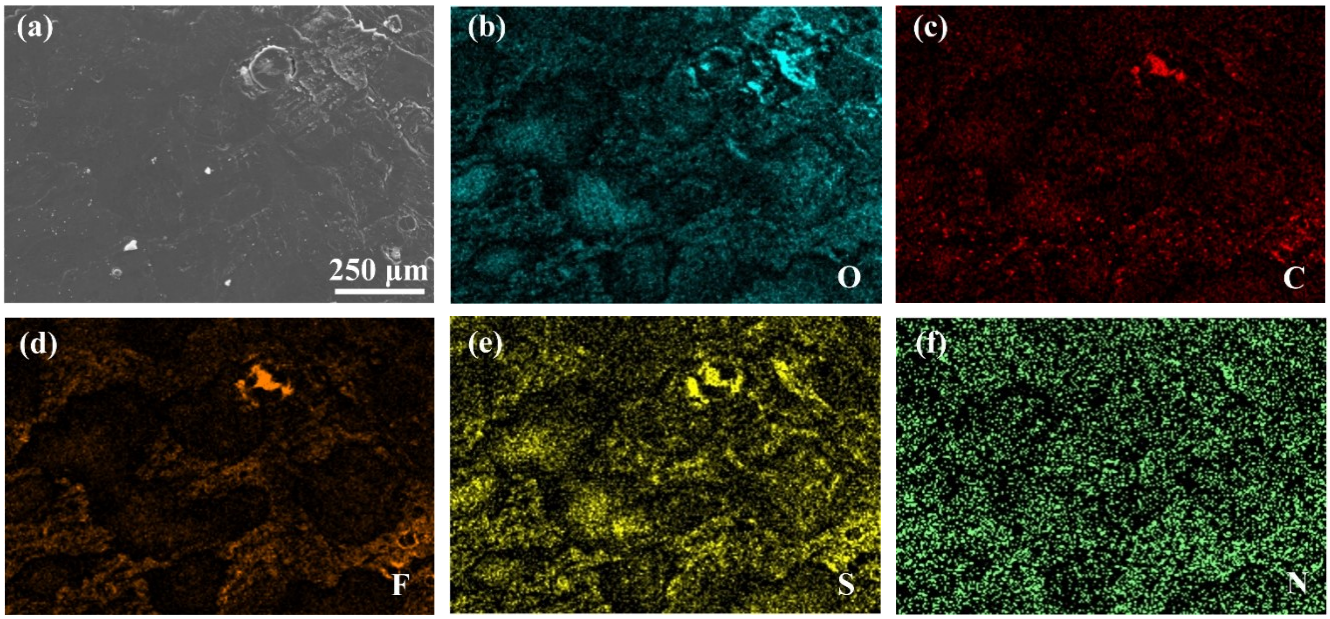


Fig. S10 Surface SEM image and corresponding EDS mapping of elemental distribution of cycled Li surface in Li|PVDF-5BFO|Li battery, (a) surface SEM image, (b) O element, (c) C element, (d) F element, (e) S element, (f) N element

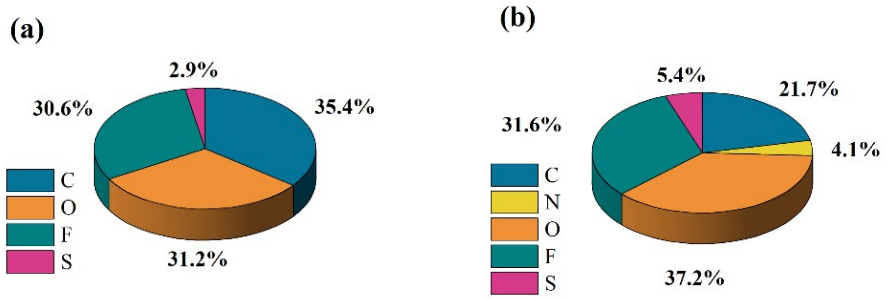


Fig. S11 Relative amount of different elements originated from the EDS analysis of the cycled Li surfaces in different batteries, (a) Li|PVDF|Li battery from Fig. S9, (b) Li|PVDF-5BFO|Li battery from Fig. S10

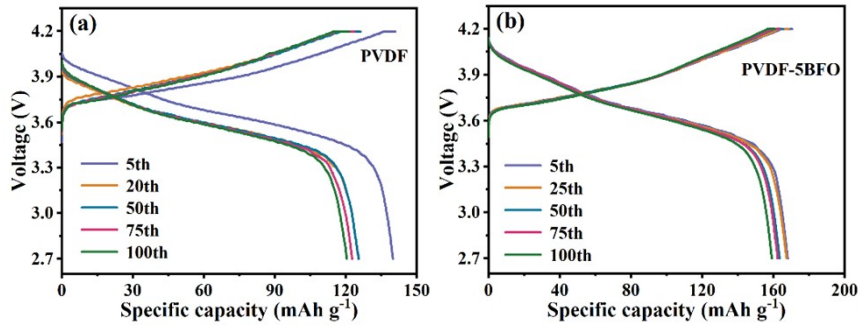


Fig. S12 Charge-discharge voltage profiles after different cycles at 0.1C, (a) Li|PVDF|NCM811 battery, (b) Li|PVDF-5BFO|NCM811 battery

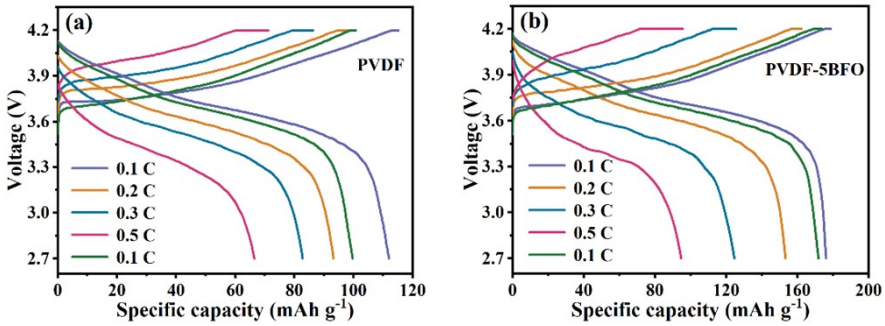


Fig. S13 Charge-discharge voltage profiles at different C-rates, (a) Li|PVDF|NCM811 battery, (b) Li|PVDF-5BFO|NCM811 battery

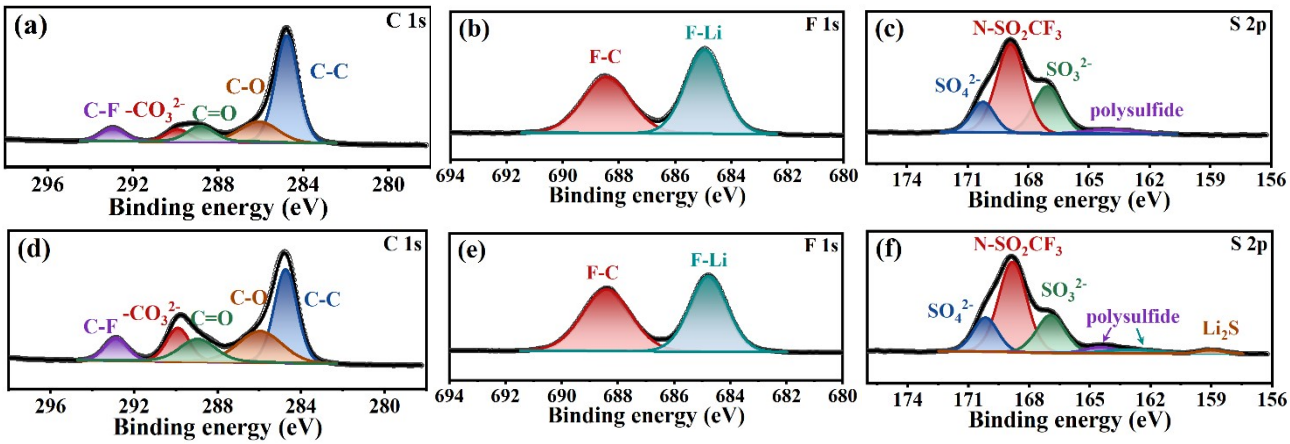


Fig. S14 XPS analysis of cycled Li metal in Li|CPE|NCM811batteries, (a) C 1s, (b) F 1s, and (c) S 2p XPS spectra of cycled Li anode from Li|PVDF| NCM811 battery, (d) C 1s, (e) F 1s, and (f) S 2p XPS spectra of cycled Li anode from Li|PVDF-5BFO|NCM811 battery

Table S1 Cycling performance of PVDF-based electrolytes incorporated different inorganic fillers

Filler tape	Cathode	C rate	Specific capacity	Cycle number	Capacity retention	Temperature	Ref.
LALZO	NCM811	0.1	160.92	100	92 %	RT	S1
LALZO	NCM811	0.2	144.70	100	85%	RT	S1
V_2O_5	NCM811	0.5	178.50	280	70%	RT	S2
LLZTNO	NCM811	0.3	137.10	80	-	RT	S3
LATP	NCM622	0.5	150.00	500	80%	30	S4
LLTO/PPC	NCM622	0.5	168.00	100	84%	RT	S5
APTES-LATP	NCM622	0.5	118.00	300	84%	40	S6
$(Mg,Al)_2Si_4O_{10}(OH)$	NCM111	0.3	121.40	200	97%	RT	S7
Li_3InCl_6	$LiFePO_4$	0.1	161.80	200	82%	RT	S8
LALZO/h-BN	$LiFePO_4$	0.2	131.00	100	92%	RT	S9
$BiFeO_3$	NCM811	0.1	168.65	100	94%	RT	This work
$BiFeO_3$	NCM811	0.3	144.89	400	89%	RT	This work

RT: room temperature (25 °C)

References

[S1] S. L. Beshahwured, Y. S. Wu, S. H. Wu, W. C. Chien, R. Jose, S. J. Lue and C. C. Yang, *Electrochim. Acta*, 2021, **366**, 137348.

[S2] T. Feng, Y. B. Hu, L. Xu, J. Q. Huang, S. B. Hu, L. F. Zhang and L. L. Luo, *Materials Today Energy*, 2022, **28**, 101062.

[S3] X. Song, T. H. Zhang, S. F. Huang, J. S. Mi, Y. D. Zhang, J. Travas-Sejdic, A. P. Turner, W. Gao and P. Cao, *J. Power Sources*, 2023, **564**, 232849.

[S4] L. Liu, D. C. Zhang, J. W. Zhao, J. D. Shen, F. K. Li, Y. Yang, Z. B. Liu, W. X. He, W. M. Zhao and J. Liu, *ACS Applied Energy Materials*, 2022, **5**, 2484-2494.

[S5] L. Zhu, H. B. Xie, W. J. Zheng and K. Zhang, *Electrochim. Acta*, 2022, **435**, 141384.

[S6] G. M. Overhoff, M. Y. Ali, J. P. Brinkmann, P. Lennartz, H. Orthner, M. Hammad, H. Wiggers, M. Winter and G. Brunklaus, *ACS Appl. Mater. Interfaces*, 2022, **14**, 53636-53647.

[S7] P. C. Yao, B. Zhu, H. W. Zhai, X. B. Liao, Y. X. Zhu, W. H. Xu, Q. Cheng, C. Jayyosi, Z. Li, J. Zhu, K. M. Myers, X. Chen and Y. Yang, *Nano Lett.*, 2018, **18**, 6113-6120.

[S8] H. Li, G. Du, H. Liang, Q. Su, D. Han, W. Zhao, M. Zhang, S. Ding and B. Xu, *J. Alloys Compd.*, 2023, **969**, 172418.

[S9] J. H. Kim, D. H. Park, J. S. Jang, J. H. Shin, M. C. Kim, S. B. Kim, S. H. Moon, S. N. Lee and K. W. Park, *Chem. Eng. J.*, 2022, **446**, 137035.