Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting Information

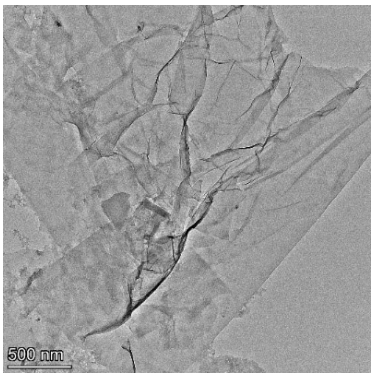

Fig. S1. TEM image of the BP nanosheets

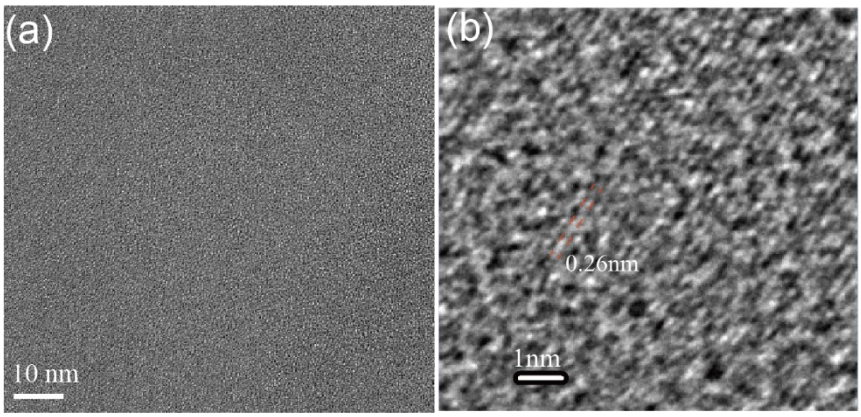

Fig. S2. High-resolution TEM images of BP nanosheets.

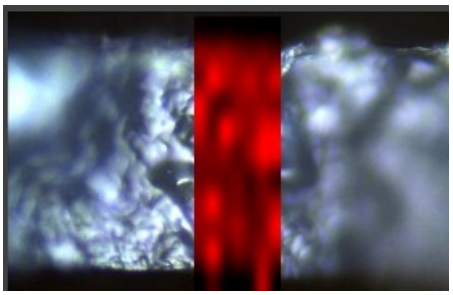

Fig. S3. The cross-sectional Raman mapping of BP SPEs.

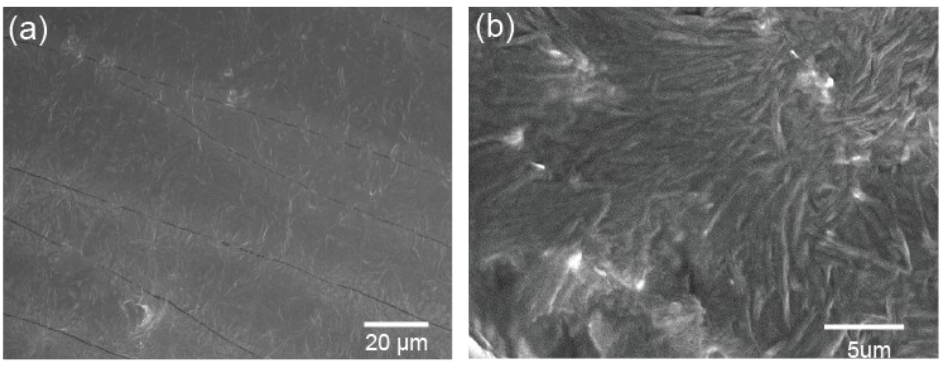

Fig. S4. The SEM images of C-BP SPEs.

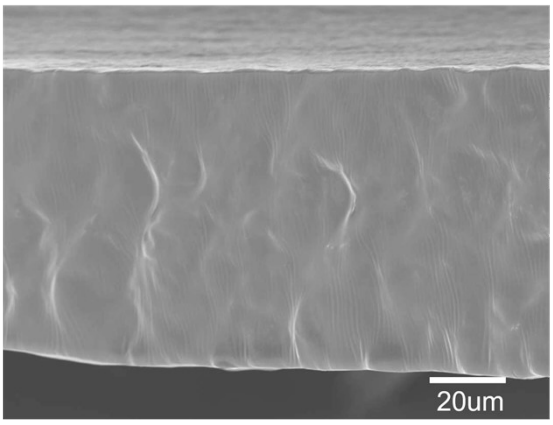

Fig. S5. The cross-section SEM images of C-BP SPEs.

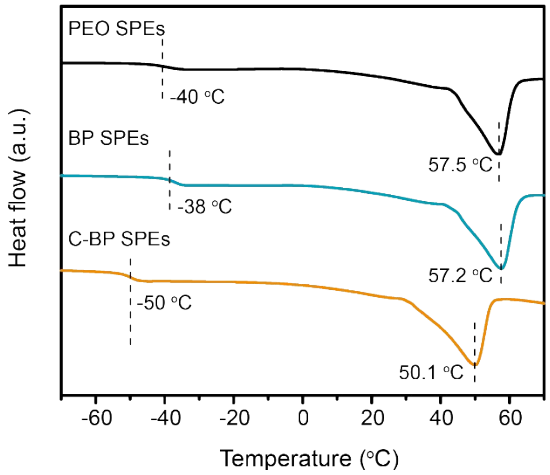

Fig. S6. The DSC curves of the PEO SPEs and C-BP SPEs from -70 to 70 °C.

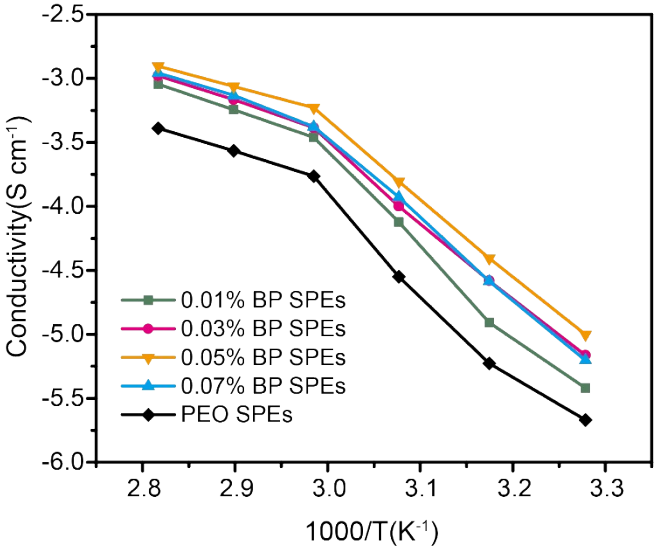

Fig. S7. BP nanosheets content-dependent ionic conductivities of SPEs at different temperatures.

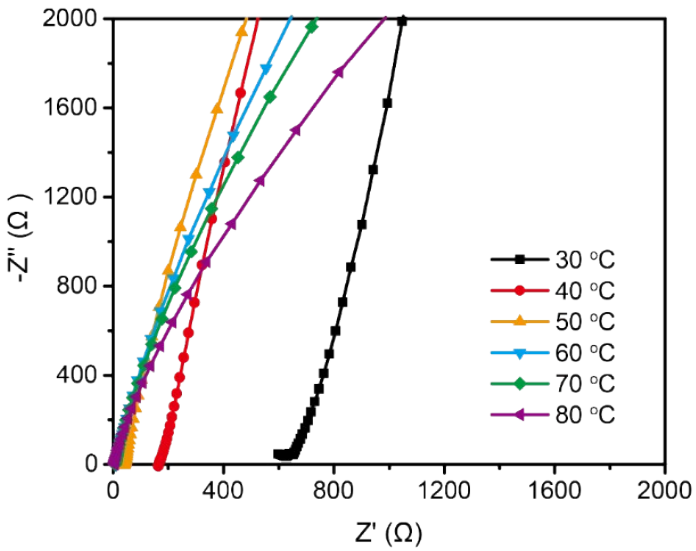

Fig. S8. EIS spectra of the BP SPEs at different temperatures.

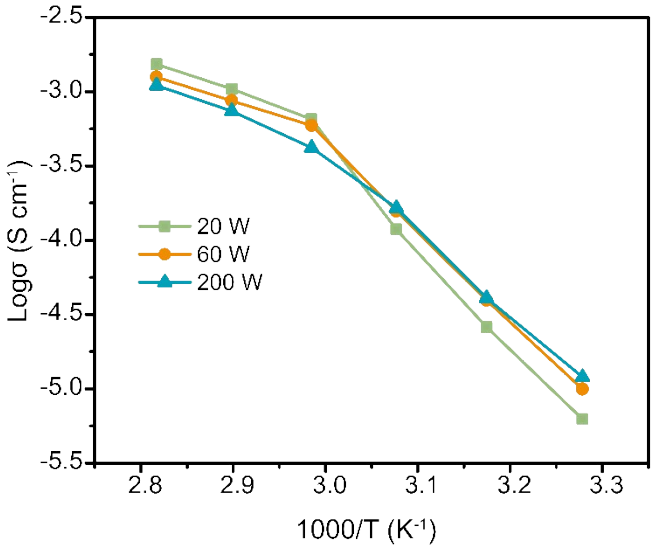

Fig. S9. Temperature dependence on the ionic conductivities of PEO SPEs with the Mw=20 w, 60 w and 200 w, respectively.

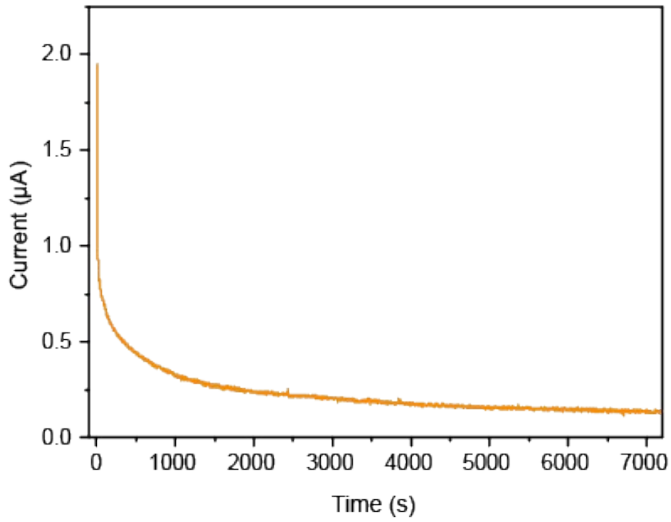

Fig. S10. Direct current polarization tests to measure the electronic conductivity of C-BP SPEs at 1 V.

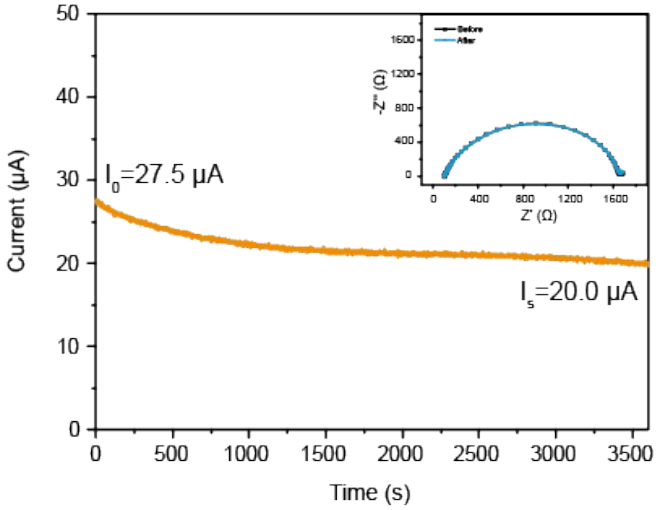

Fig. S11. Chronoamperometry profiles and EIS spectra (inset) obtained before and after polarization for the symmetric Li/SPEs/Li cells prepared with BP SPEs.

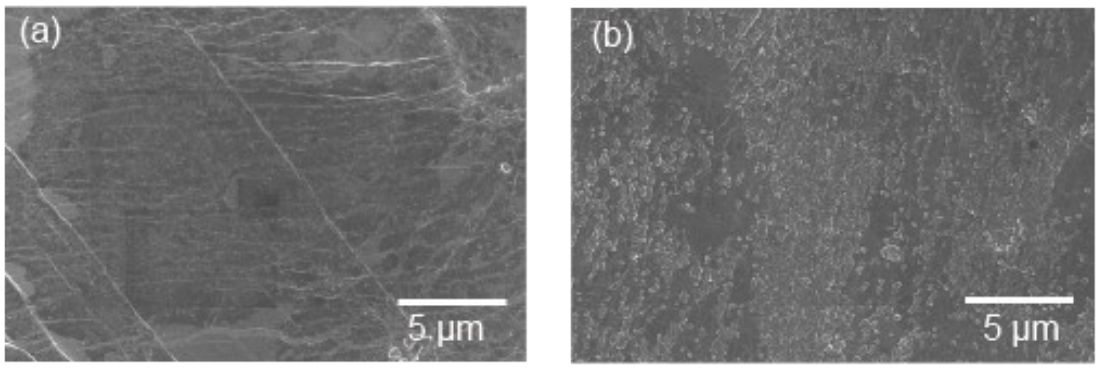

Fig. S12. the SEM image of Lithium metal with C-BP SPEs (a) and with PEO SPEs (b) after 100 cycles.

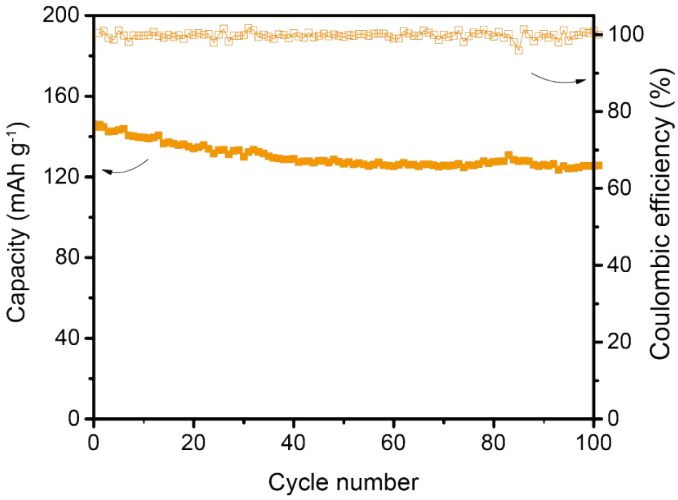

Fig. S13. Cycle performance of NCM622/SPEs/Li batteries using C-BP SPEs at 0.1 C and 30 °C

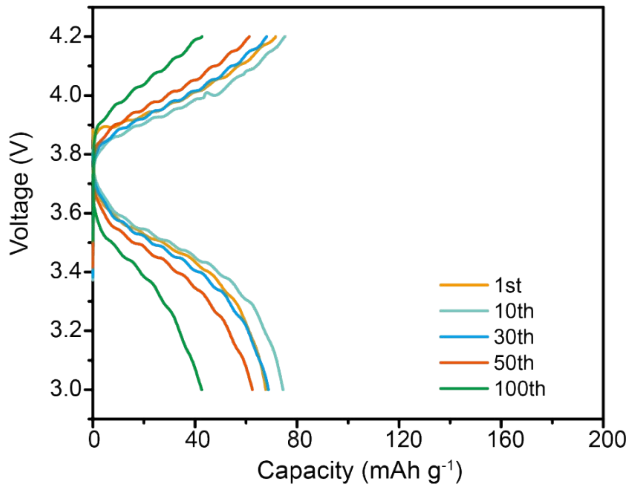

Fig. S14. The charge and discharge profiles of the 1st, 10th, 30th, 50th and 100th cycles of the NCM622/SPEs/Li cells prepared using PEO SPEs at 0.2 C and RT.

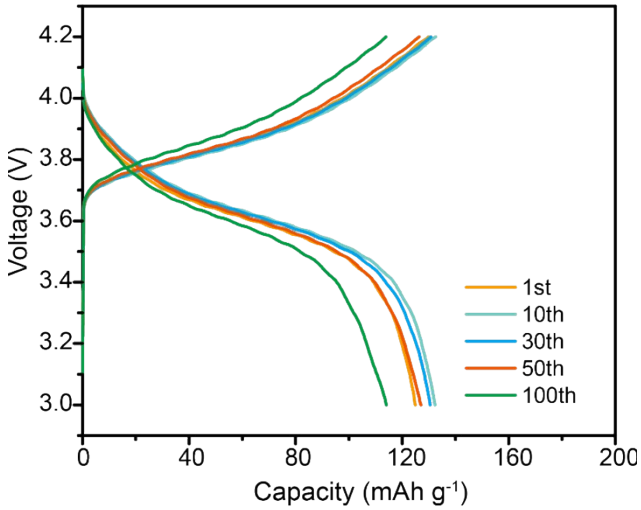

Fig. S15. The charge and discharge profiles of the 1st, 10th, 30th, 50th and 100th cycles of the NCM622/SPEs/Li cells prepared using C-BP SPEs at 0.2 C and RT.

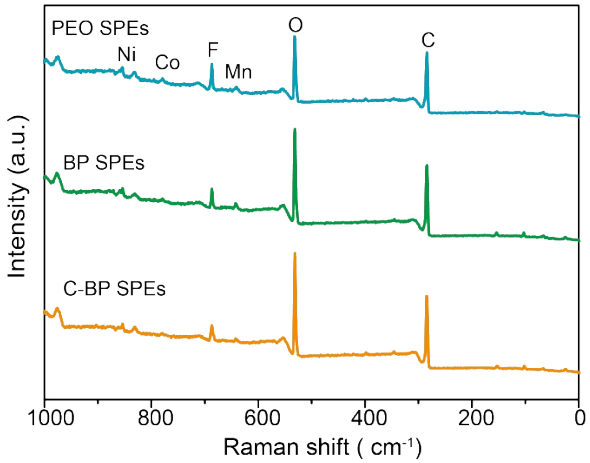

Fig. S16. The XPS spectra of the NCM622 cathodes using PEO SPEs, BP SPEs and C-BP SPEs after 5 cycles.

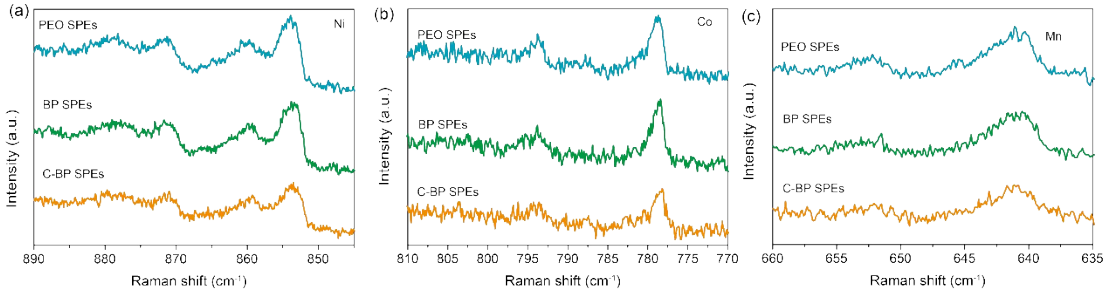

Fig. S17. The XPS spectra of (a) Ni 2p, (b) Co 2p, (c) Mn 2p of the NCM622 cathodes using PEO SPEs, BP SPEs and C-BP SPEs after 5 cycles.

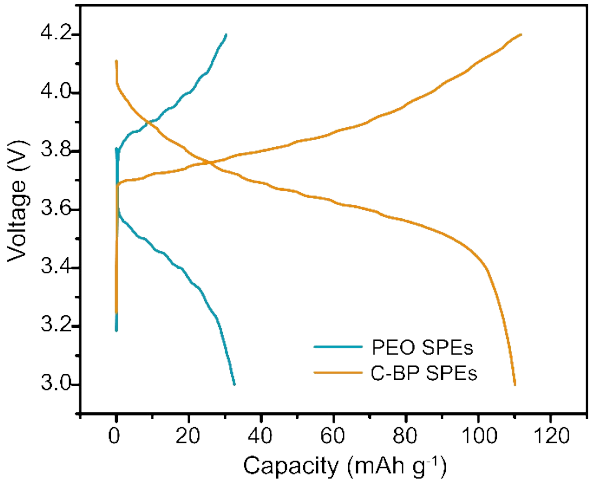

Fig. S18. The charge and discharge curves of the NCM622/SPEs/Li pouch cells using the PEO SPEs, and C-BP SPEs at 0.1 C

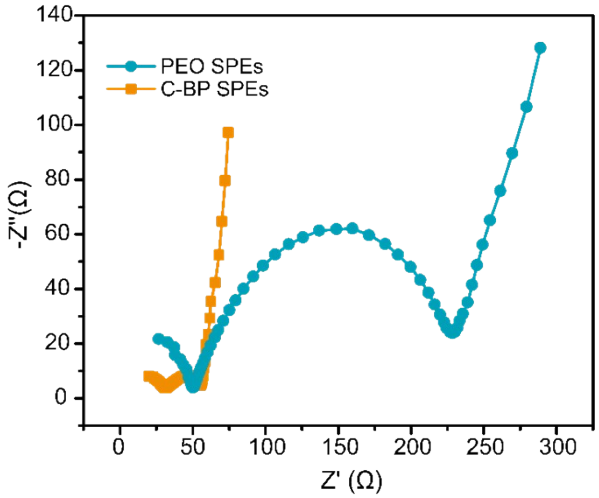

Fig. S19. EIS of the pouch cells prepared using PEO SPEs and C-BP SPEs at 30 °C

Table S1. Comparisons of electrochemical performances of PEO SPEs with the addition of different kinds of 2D nanosheets.

Solid electrolyte	**The mass content of nanofiller**	**Working temperature (°C)**	**Voltage (V)**	**Discharge capacity (mAh g^{-1})**	**Capacity retention**	**cathode**	**Ref.**
PEO/LiTFSI/ox-PIL@GO	5%	40	2.5-4.0	116	95% (1C,300cycles)	LFP	[1]
PEO/LiTFSI/5% MnO2 CSPE	5%	60	2.5-4.0	163.8	87.6% (0.5C 300cycles)	LFP	[2]
PE-hG	1%	80	2.4-3.8	135.8	71% (1C after 300 cycles)	LFP	[3]
PEO/LiTFSI/10% VS SPE	10%	60	2.5-3.8	160	NONE	LFP	[4]
VAVS–CSPE	10%	30	2.5-3.8	167 (0.1 C) 135 (0.5 C)	88% (0.1 C)	LFP	[5]
5% g-C3N4 CSPE	5%	60	2.8-3.8	161.3	96% (0.2 C after 100 cycles)	LFP	[6]
C-BP SPEs	0.05%	30	3-4.2	150.2 (0.1 C) 134.3 (0.2 C) 105.1 (0.5 C)	88.1% (0.5 C after 500 cycles)	NCM622	This work

Reference

1. W. Bao, Z. Hu, Y. Wang, J. Jiang, S. Huo, W. Fan, W. Chen, X. Jing, X. Long and Y. Zhang, *Chem. Eng. J.*, 2022, **437**, 135420.
2. Y. Li, Z. Sun, D. Liu, Y. Gao, Y. Wang, H. Bu, M. Li, Y. Zhang, G. Gao and S. Ding, *J. Mater. Chem. A*, 2020, **8**, 2021.
3. J. Xu, Y. Meng, Q. Ding, R. Wang, T. Gan, J. Zhang, Z. Lin and J. Xu, *J. Mater. Chem. A*, 2022, **10**, 4402.
4. W. Tang, S. Tang, C. Zhang, Q. Ma, Q. Xiang, Y. W. Yang and J. Luo, *Adv. Energy Mater.*, 2018, **8,** 1800866.
5. W. Tang, S. Tang, X. Guan, X. Zhang, Q. Xiang and J. Luo, *Adv. Funct. Mater.*, 2019, **29**, 1900648.
6. Z. Sun, Y. Li, S. Zhang, L. Shi, H. Wu, H. Bu and S. Ding, *J. Mater. Chem. A*, 2019, **7**, 11069.