

Supplementary Materials for

Gradient-integrated bilayer solid polymer electrolyte enabling enhanced room-temperature cyclability for rechargeable lithium metal batteries

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Supplementary Figures

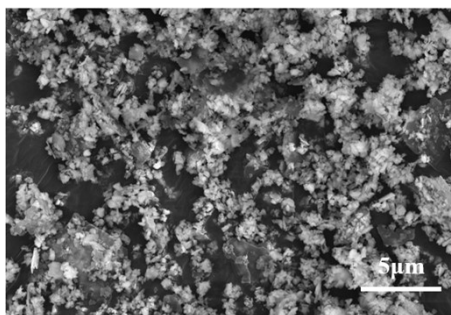


Fig.S1 SEM image of the commercial LLZTO particle

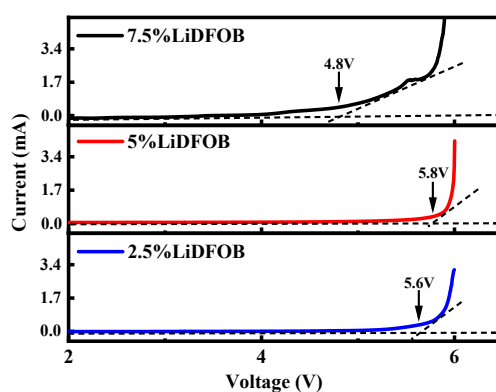


Fig.S2 LSV curves of the PVDF layer with different LiDFOB amounts

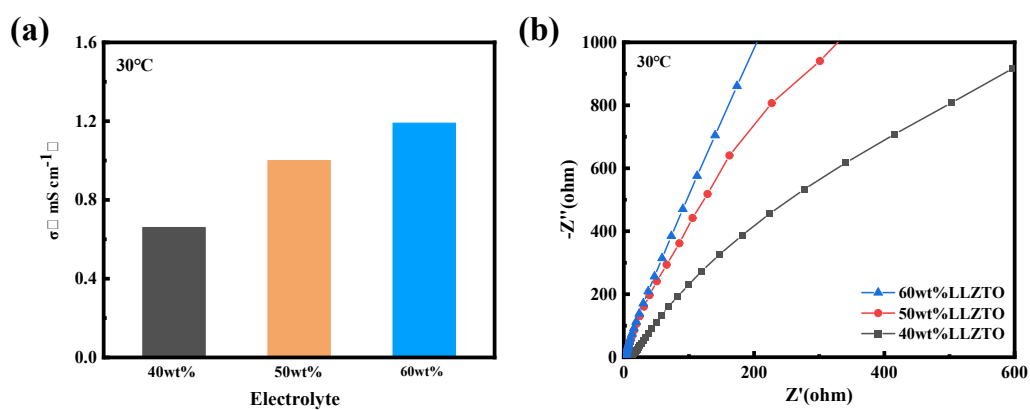


Fig.S3 (a) Ionic conductivities and (b) EIS plots of the PEO layers with various LLZTO contents

at 30 °C

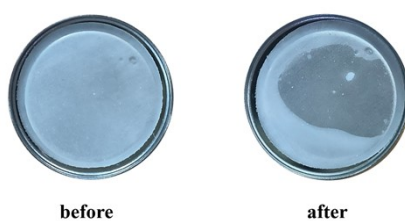


Fig. S4 Pictures of the PVDF layer assembled in the coin cell before and after external pressure

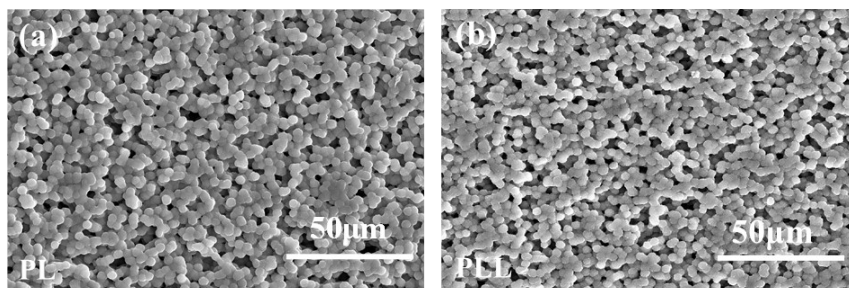


Fig.S5 SEM images of (a) PL and (b) PLL electrolytes

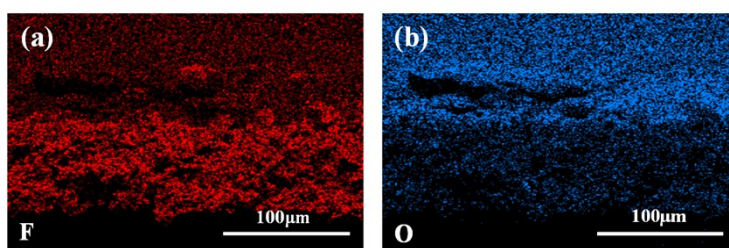


Fig.S6 The EDS mappings of (a) F element and (b) O element corresponding to the cross-section image of the g-BSPE in Fig.2c

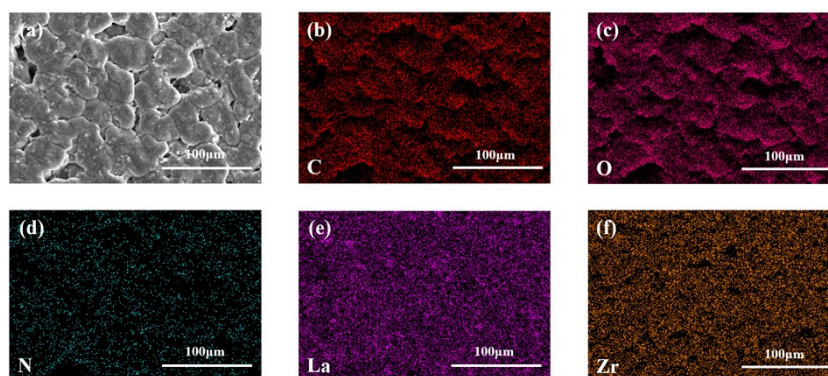


Fig.S7 Surface image of (a) the PEO layer and (b) the corresponding EDS mappings for elemental distribution analyses

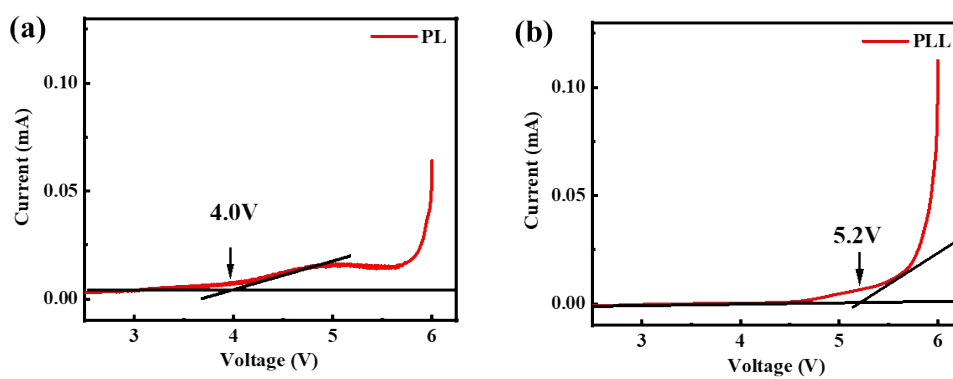


Fig.S8 LSV curves of (a) PL and (b) PLL electrolytes

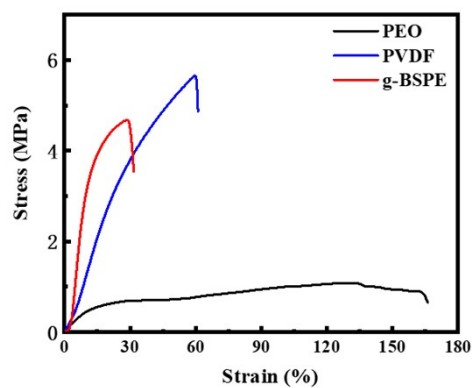


Fig.S9 Stress-strain curves of PEO layer, PVDF layer and g-BSPE

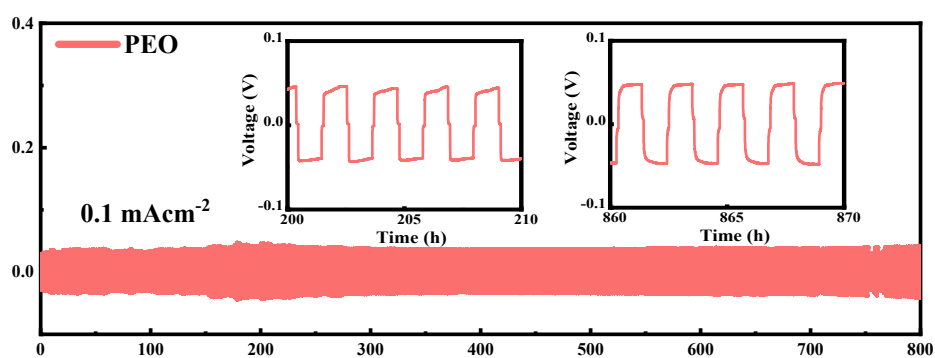


Fig.S10 Plating/stripping cycling of Li|Li symmetrical cells in the PEO layer at 0.1 mA cm⁻²

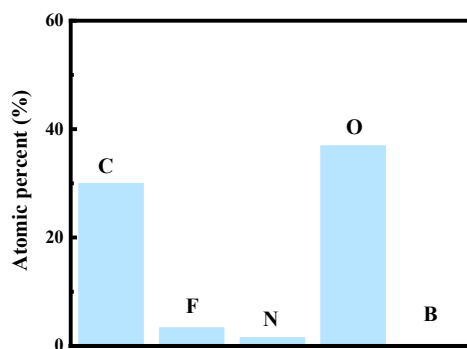


Fig.S11 The atomic percent on the Li surface in contact with PEO layer after cycling

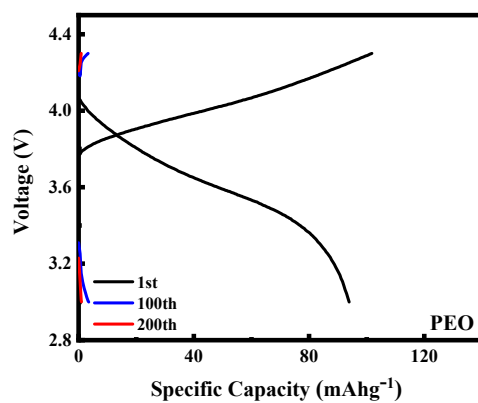


Fig.S12 Charge/discharge profiles of the NCM/Li cell in the PEO layer during cycling at 1C

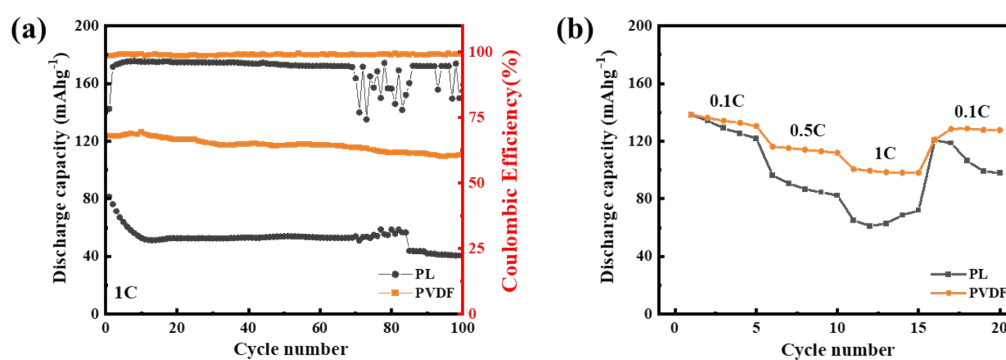


Fig.S13 Comparison of (a) cycle and (b) rate performance between NCM/PVDF/Li cell and NCM/PL/Li cell

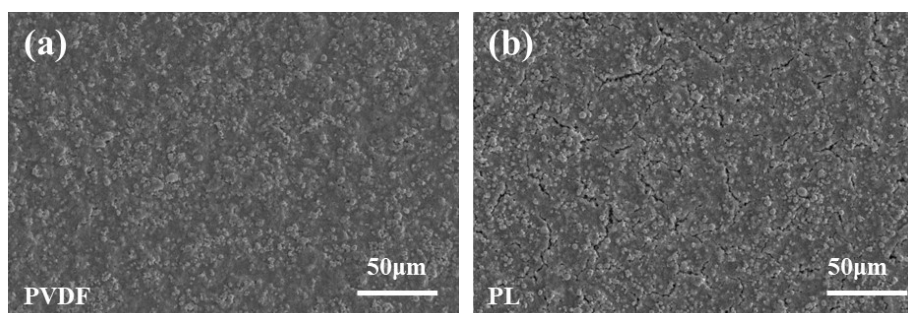


Fig.S14 SEM images of the cycled cathodes from (a) NCM/PVDF/Li cell and (b) NCM/PL/Li cell

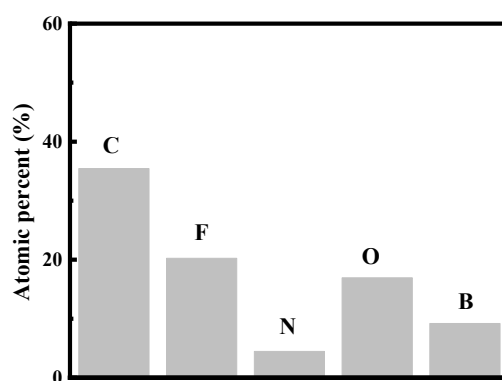


Fig.S15 The atomic percent on the NCM cathode after cycling in the g-BSPE

Supplementary Tables

Table S1 Particle size distribution of the commercial LLZTO

	D10	D50	D90	D100
LLZTO	$\geq 200\text{nm}$	$\approx 400\text{ nm}$	$\leq 800\text{ nm}$	$\leq 1000\text{ nm}$

Table S2 Ionic conductivities of various SPEs at different temperatures

SPE	Ionic conductivity (σ) /mS cm ⁻¹				
	30°C	40°C	50°C	60°C	70°C
PEO	1.00	1.34	1.63	1.83	2.00
PVDF	0.64	0.94	1.14	2.70	2.94
g-BSPE	0.99	1.37	1.61	2.09	2.33
PL	0.13	0.14	0.23	0.31	0.40
PLL	0.60	0.78	0.81	1.10	1.61