

Supporting Information

Stable and Flexible Sulfide Composite Electrolyte for High-Performance Solid-State Lithium Batteries

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Table S1. Comparison of ionic conductivities of composite electrolytes with different polymers and solid electrolytes.

	Composite electrolyte	Solid electrolyte (wt%)	Ionic conductivity (S cm⁻¹)	Refs.
Sulfide	PVDF-HFP/Li ₇ PS ₆	10	1.11×10 ⁻⁴	This work
	PEO/Li ₁₀ GeP ₂ S ₁₂	1	1.18×10 ⁻⁵	37
Oxide	PEO/Li _{0.3} La _{0.557} TiO ₃	—/—	1.8×10 ⁻⁴	23
	PAN/Li _{0.3} La _{0.557} TiO ₃ NWs	15	2.4×10 ⁻⁴	24
	PEO/AAO	—/—	1.79×10 ⁻⁴	25
	PEO/Li _{1.3} Al _{0.3} Ti _{1.7} (PO ₄) ₃	—/—	1.6×10 ⁻³ (80 °C)	26
	PVDF/Li _{6.75} La ₃ Zr _{1.75} Ti _{0.25} O ₁₂	10	5.0×10 ⁻⁴	27
	PVDF-HFP/Li ₇ La ₃ Zr ₂ O ₁₂	50	1.1×10 ⁻⁴	28
	PEO/Li ₇ La ₃ Zr ₂ O ₁₂ NWs	—/—	2.39×10 ⁻⁴	29
	PEO/Li ₇ La ₃ Zr ₂ O ₁₂	40	5×10 ⁻⁵ (80 °C)	30
	PEO/Li ₇ La ₃ Zr ₂ O ₁₂	52.5	~1.0×10 ⁻⁵ (35 °C)	31
	PEO/Li _{6.4} La ₃ Zr ₂ Al _{0.2} O ₁₂ NWs	—/—	2.5×10 ⁻⁴	32
	PEO/Li _{1.5} Al _{0.5} Ge _{1.5} (PO ₄) ₃	20	6.67×10 ⁻⁴ (60 °C)	33
	PEO/Li _{1.5} Al _{0.5} Ge _{1.5} (PO ₄) ₃	70	1.0×10 ⁻⁵	34
	PEO/Li _{1.5} Al _{0.5} Ge _{1.5} (PO ₄) ₃	99	—/—	35
	PVDF/Li _{1.3} Al _{0.3} Ti _{1.7} (PO ₄) ₃	33.3	9.6×10 ⁻⁴	36

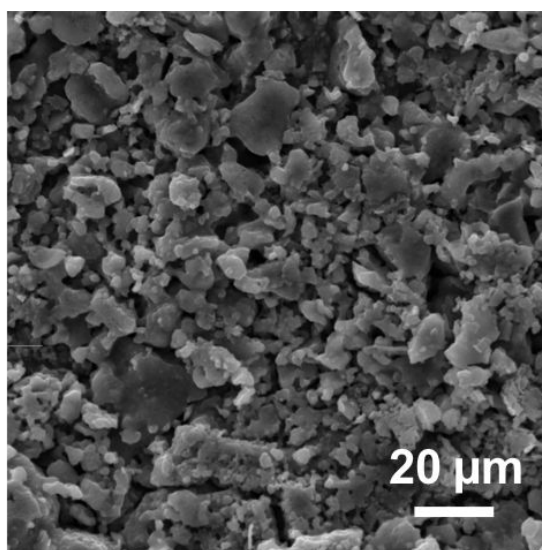


Figure S1. SEM image of Li_7PS_6 SE. The particle size of Li_7PS_6 is around 3–5 μm .

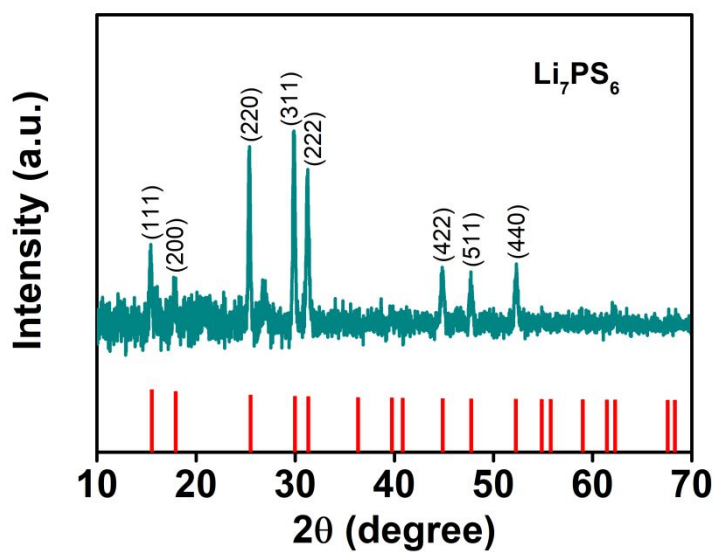


Figure S2. XRD pattern of Li_7PS_6 SE. This is in good agreement with the cubic structure of $F-43m$ symmetry.

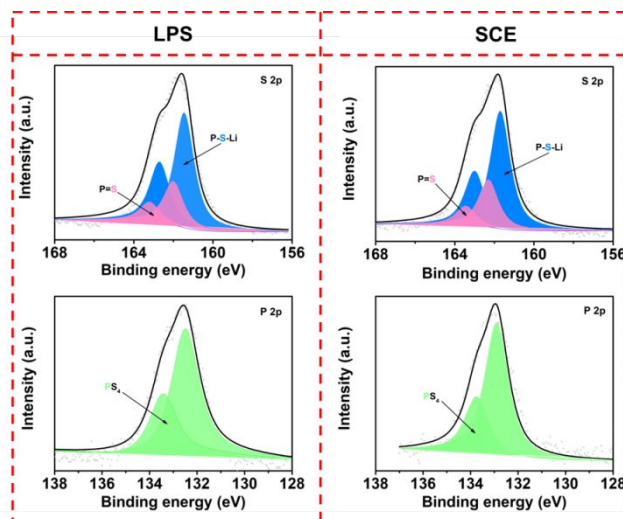


Figure S3. XPS spectra of S 2p and P 2p for pure LPS and SCE.

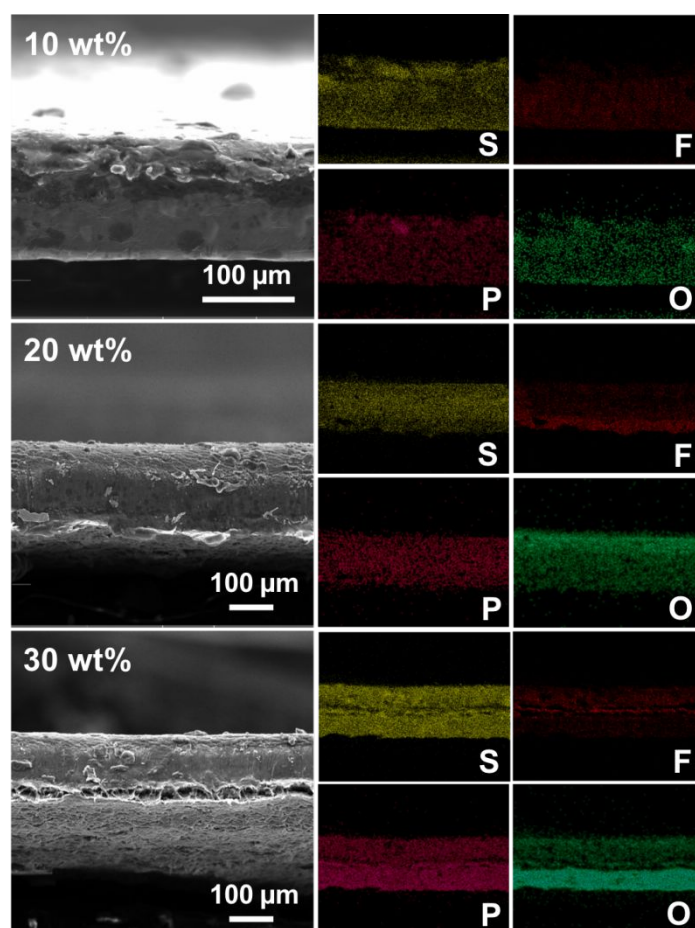


Figure S4. Cross-sectional SEM images and EDS mappings of SCE membranes with various contents of LPS (10, 20 and 30 wt%).

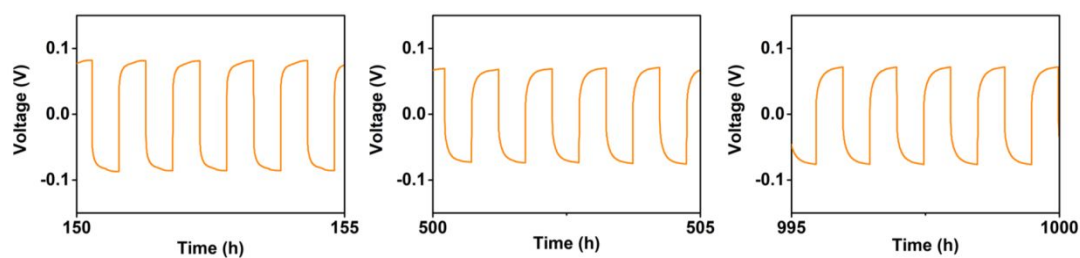


Figure S5. Zoomed-in voltage profiles for typical cycles of Li symmetric cell with SCE at current density of 0.2 mA cm^{-2} .

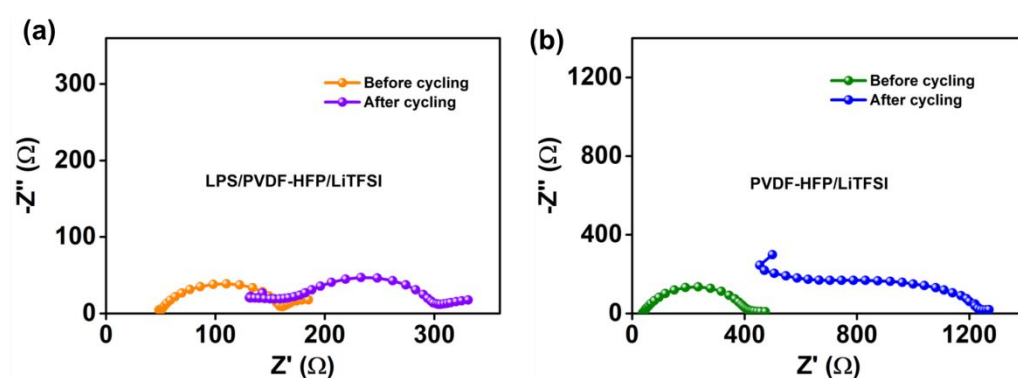


Figure S6. EIS of symmetric Li cells after cycling with (a) LPS/PVDF-HFP/LiTFSI SCE after 180 cycles and (b) PVDF-HFP/LiTFSI polymer electrolyte after 100 cycles.

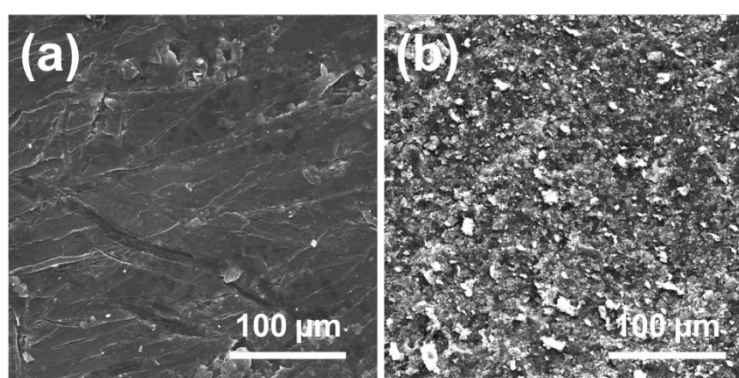


Figure S7. SEM images of the Li metal surfaces for (a) LPS/PVDF-HFP/LiTFSI SCE and (b) PVDF-HFP/LiTFSI polymer electrolyte after cycling.

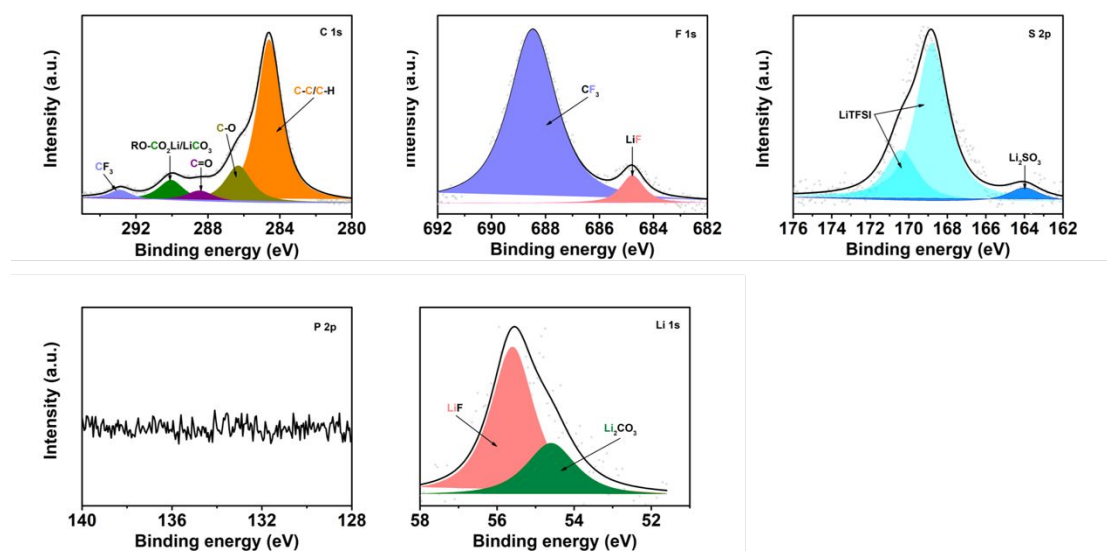


Figure S8. Detailed XPS spectra of C 1s, F 1s, S 2p, P 2p and Li 1s obtained from Li metal surface of LFP/Li cell with PVDF-HFP/LiTFSI after cycling.

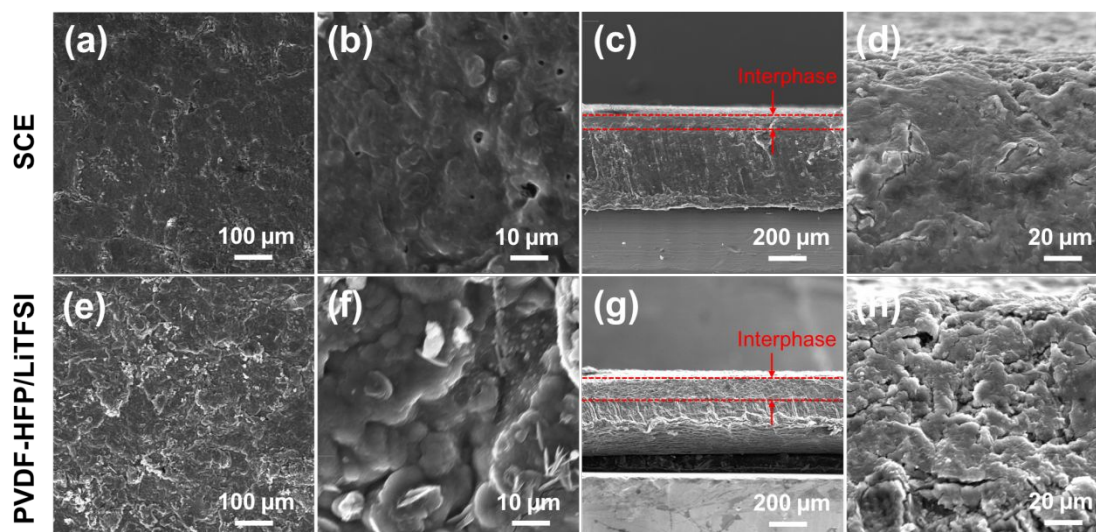


Figure S9. SEM images of surface and cross-section for the cycled Li anodes from LFP/Li cells assembled with (a–d) SCE and (e–h) PVDF-HFP/LiTFSI.