

Supporting Information

Self-densified ultrathin solid electrolyte membrane fabricated from monodispersed sulfide electrolyte nanoparticles

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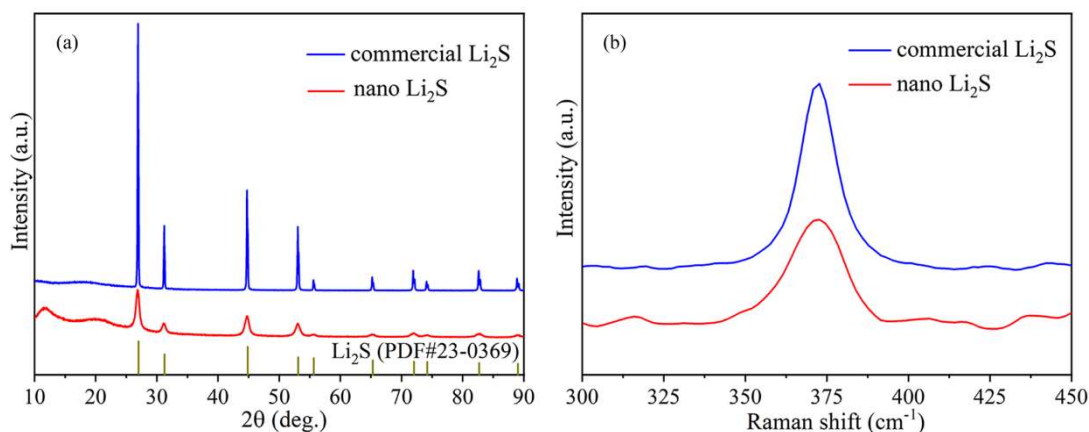


Figure S1 (a) XRD patterns (b) Raman spectra of the as-synthesized nano- Li_2S and commercial Li_2S .

Peak broadening is evidently observed for nano- Li_2S , reflecting its significantly reduced dimension.

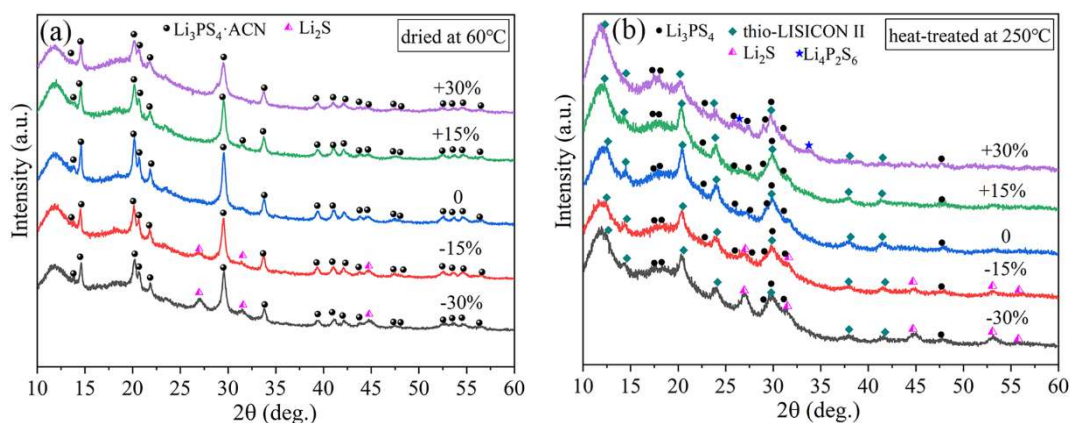


Figure S2 XRD patterns of the LPS samples synthesized by different proportions of $\text{Li}_2\text{S}\cdot\text{P}_2\text{S}_5$ and Li_2S after (a) drying at 60°C and (b) heat treatment at 250°C . The symbol “0” denotes that $\text{Li}_2\text{S}\cdot\text{P}_2\text{S}_5$ and Li_2S sol are mixed at 3:1 molar ratio of Li_2S and P_2S_5 . “-” and “+” respectively represent the deficient and excess of $\text{Li}_2\text{S}\cdot\text{P}_2\text{S}_5$ on the base of “0”, and the numbers behind “-” or “+” denote the percentage of shortage or excess.

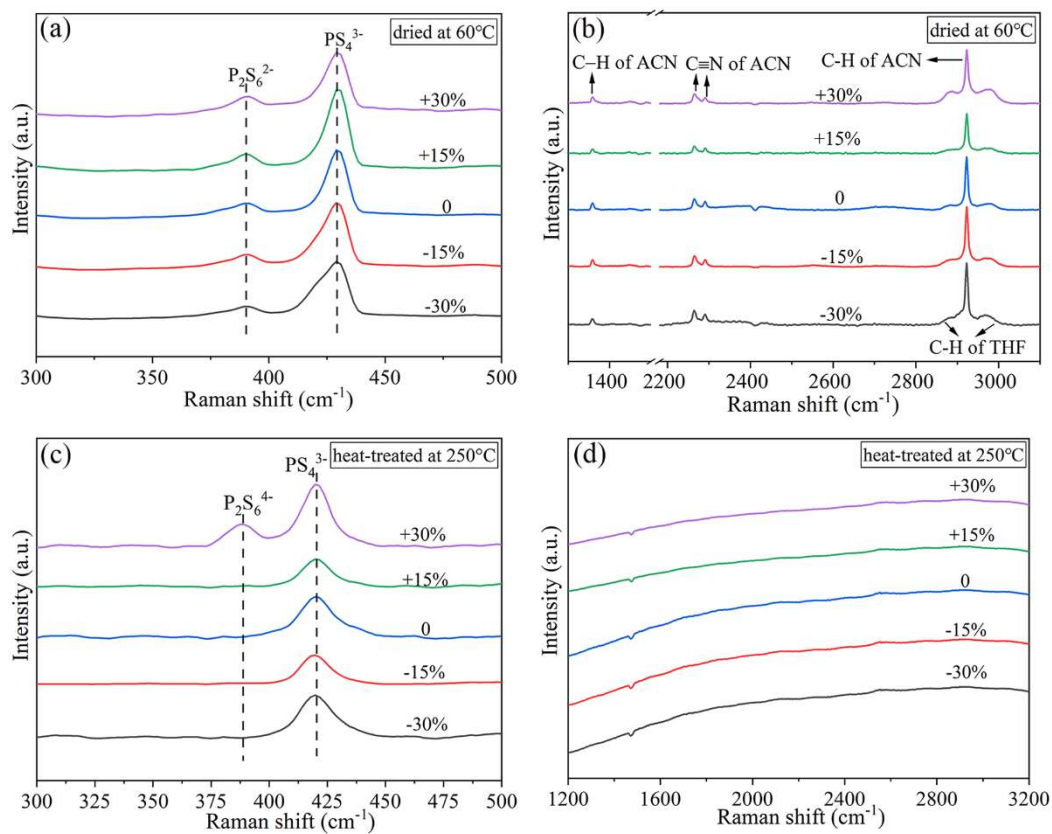


Figure S3 Raman spectra of the LPS samples synthesized by different proportions of $\text{Li}_2\text{S}\cdot\text{P}_2\text{S}_5$ and Li_2S after (a) drying at 60°C and (b) heat treatment at 250°C . Signals attributed to the solvents disappear after 250°C heat treatment, indicating the solvents are removed.

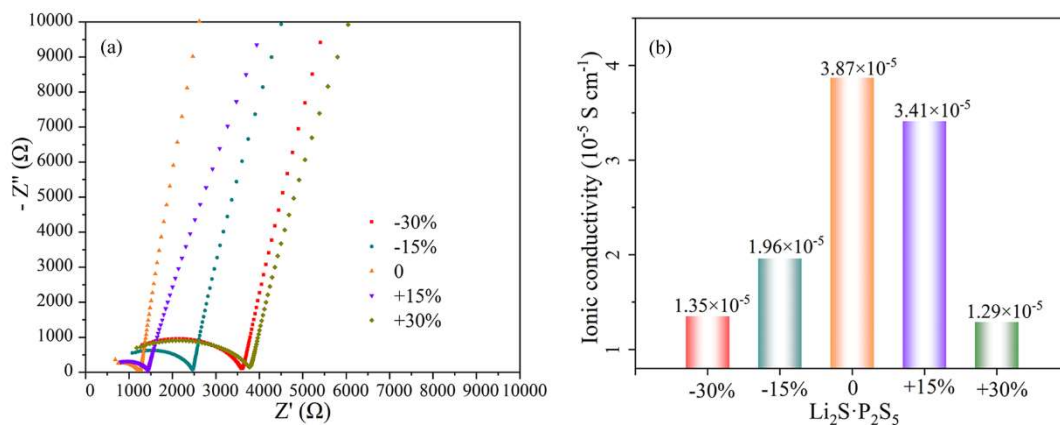


Figure S4 (a) Impedance spectra of the pelletized LPS solid electrolytes synthesized with different proportions of $\text{Li}_2\text{S}\cdot\text{P}_2\text{S}_5$ and Li_2S . (b) Room temperature ionic conductivity of the LPS solid electrolytes calculated from the impedance spectra.

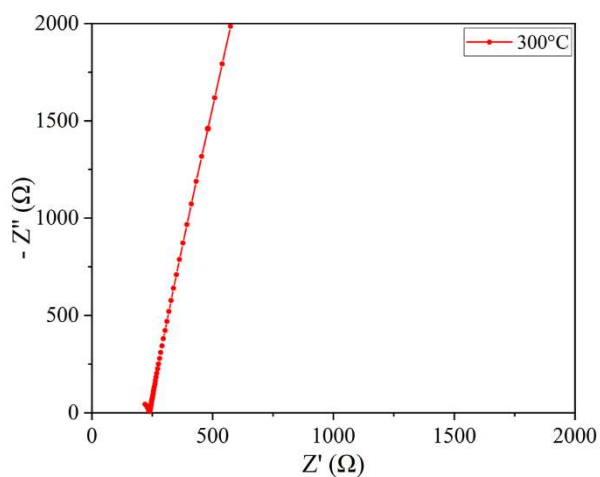


Figure S5 Impedance spectrum of the pelletized LPS solid electrolyte after heat treatment at 300°C.

The dimension of the pellet is $\Phi 15 \text{ mm} \times 0.68 \text{ mm}$, resulting in a conductivity of $1.69 \times 10^{-4} \text{ S cm}^{-1}$.

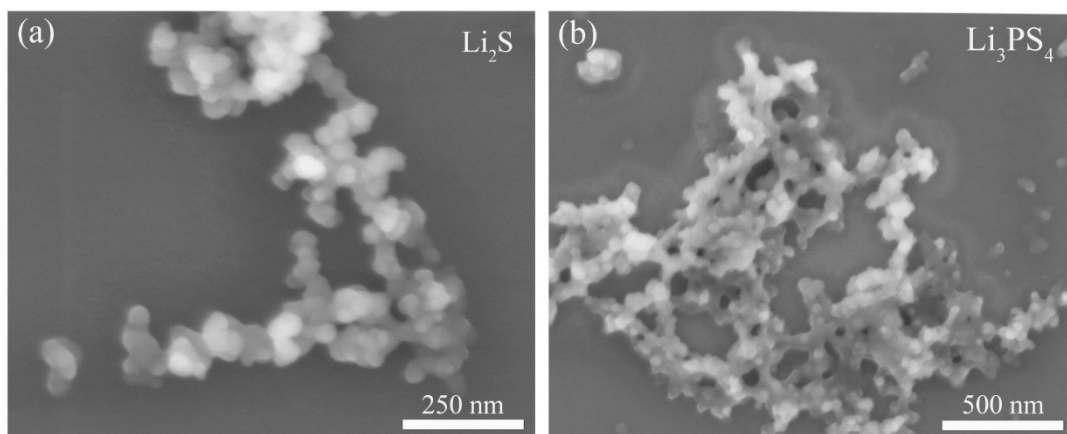


Figure S6 SEM images of (a) the nano-Li₂S prepared by mixing sulfur powder and Li-Naph solution, and (b) the resultant LPS precursor particles.

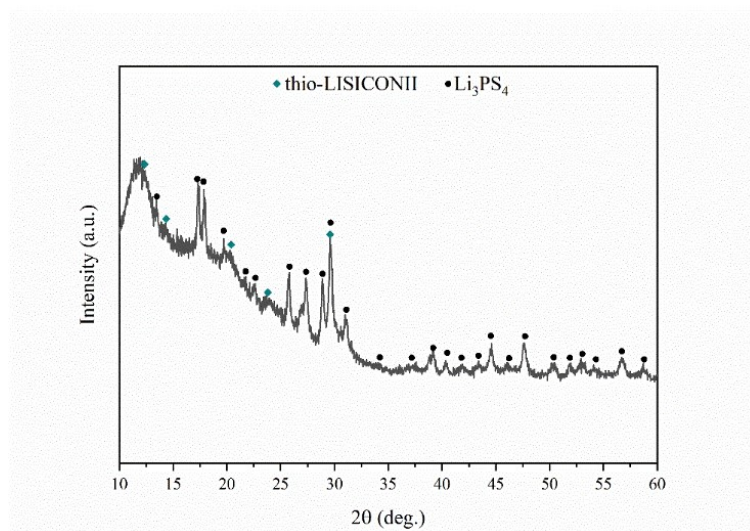


Figure S7 XRD patterns of LPS prepared by dual precursor route using the nano-Li₂S prepared by mixing sulfur powder and Li-Naph solution.

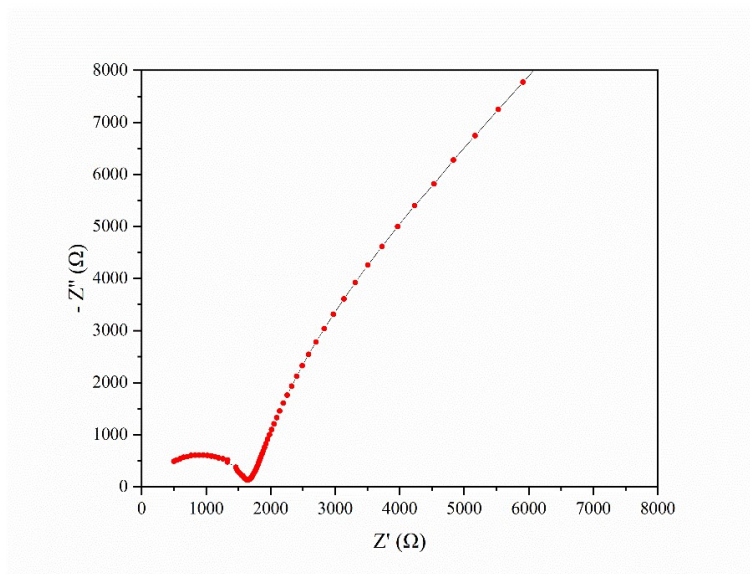


Figure S8 Impedance spectrum of the pelletized LPS solid electrolyte after heat treatment at 250°C.

The LPS is synthesized with the nano-Li₂S prepared by mixing sulfur powder and Li-Naph solution.

The dimension of the pellet is $\Phi 15 \text{ mm} \times 0.84 \text{ mm}$, resulting in a conductivity of $2.8 \times 10^{-5} \text{ S cm}^{-1}$.

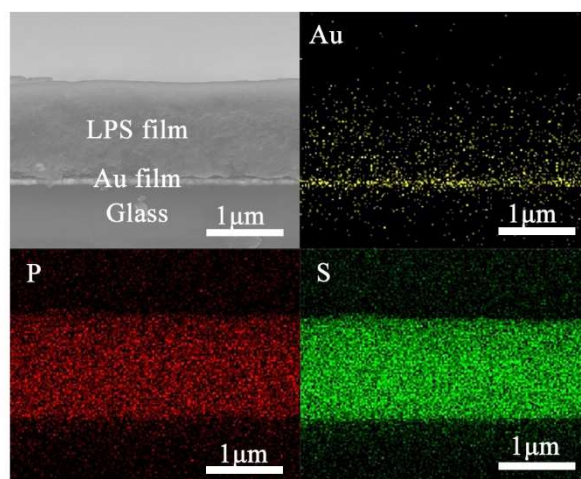


Figure S9 SEM image and EDS elemental mapping of the cross section of a LPS membrane on an Au-coated glass substrate before Li deposition.

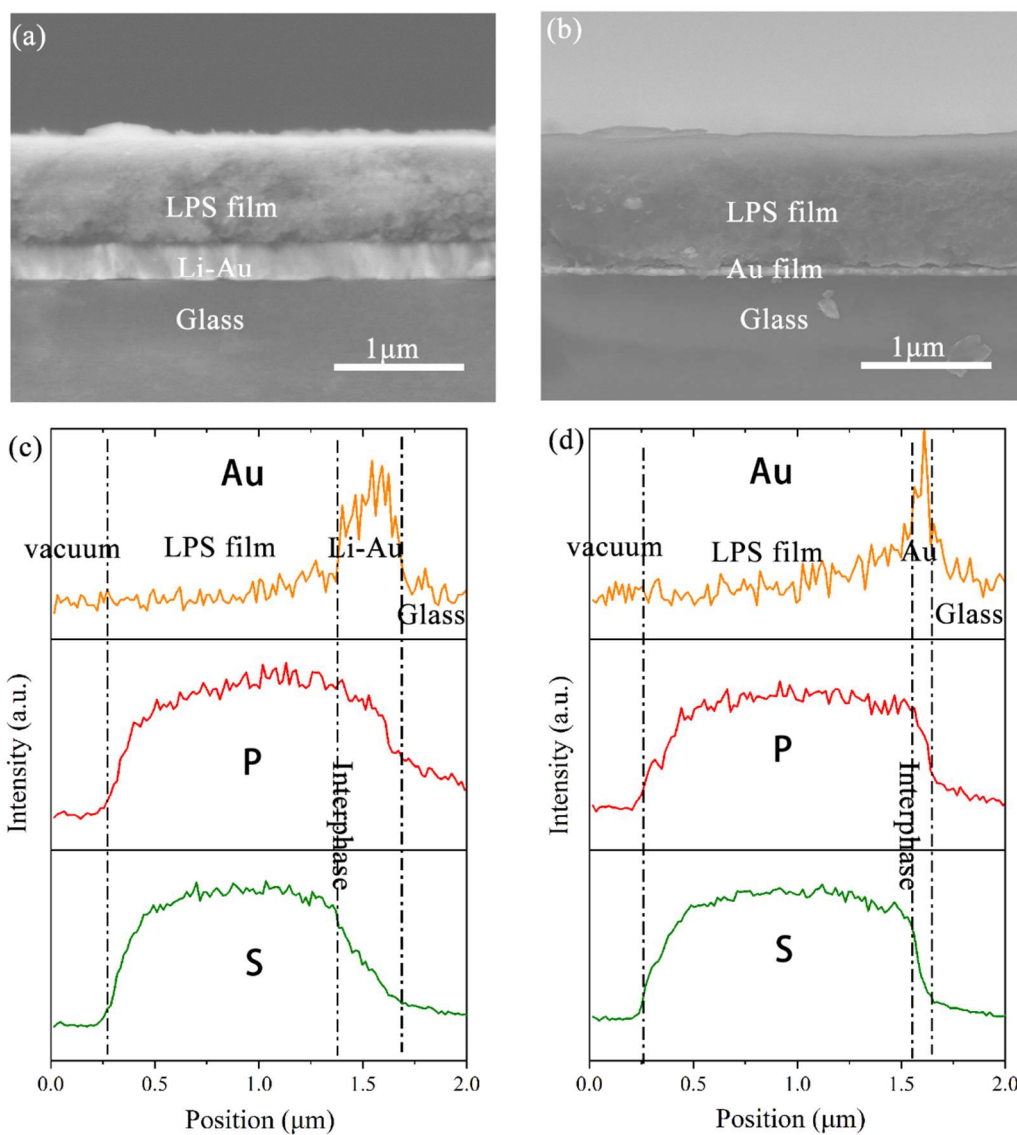


Figure S10 SEM images of a LPS membrane on an Au-coated glass substrate (a) after lithium deposition with a capacity of $46.3 \mu\text{Ah cm}^{-2}$ at a current of $65 \mu\text{A cm}^{-2}$ and (b) before Li deposition. The corresponding EDS line scanning results of the LPS membrane after and before lithium deposition are shown in (c) and (d), respectively.

Table S1 Parameters of reported sulfide electrolyte membranes.

Electrolyte	Thickness (μm)	Resistance ($\Omega \text{ cm}^2$)	Particle form	Densification processing	Ref.
Li_3PS_4	0.5–1	0.8	nanoparticles in sol precursor	spin coating + low temperature sintering	This work
Li_3PS_4	50	42	nanosheets in suspension precursor	cold pressing at 200 MPa	S1
Li_3PS_4	0.4–35	17	nanosheets in suspension precursor	hot pressing at 200 MPa	S2
$\text{Li}_6\text{PS}_5\text{Cl}$	35	18	micro-sized particles + binder	cold pressing at 150 MPa	S3
$\text{Li}_{5.4}\text{PS}_{4.4}\text{Cl}_{1.6}$	30	0.35	micro-sized particles+ binder	cold pressing at 500 MPa	S4
$\text{Li}_6\text{PS}_5\text{Cl}$	60	0.95	micro-sized particles + scaffold	cold pressing at 500 MPa	S5
Li_3PS_4	70	35	micro-sized particles + scaffold	cold pressing at 370 MPa	S6
$\text{Li}_{10}\text{GeP}_2\text{S}_{12}$	90	26	micro-sized particles + scaffold	cold pressing at 370 MPa	S6

Table S2 Assumed parameters for the energy-density estimation of a Li-S battery using LPS solid electrolyte membrane.

Average cell voltage	2.1 V Li/Li ⁺
Active material loading (cathode)	0.5–5.5 mg cm ⁻²
Percentage of active material in composite cathode	30%
Anode/cathode capacity ratio	3
Al current collector thickness	10 μm
LPS membrane thickness	1 μm or 30 μm
Density of LPS	1.868 g cm ⁻³

Battery structure and calculation method follow the works in Ref. S7.

References

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