Supporting Information for

All-Solid-State Thin-Film Lithium-Sulfur Batteries

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Supplementary Figures



Fig. S1 Potential profiles of liquid-electrolyte lithium-sulfur battery using Li_2S_6 electrolyte

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Fig. S2 Cross-section SEM image of the VGs



Fig. S3 FFT patterns, inverse FFT patterns



Fig. S4 HRTEM image of the graphene sheet.



Fig. S5 Electrochemical performance of the VGs-Li₂S thin-film cathode in liquid system. **a** CV curves; **b** voltage profiles at a current density of 25.8 μ A cm⁻²; **c** cycling performances



Fig. S6 Voltage profiles of the VGs-Li₂S thin-film cathode under different current densities



Fig. S7 Cycling performances of VGs-Li₂S/LiPON/Pre-Li cells at 10.5 μ A cm⁻² with an initial Coulombic Efficiency of 78.6%

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Fig. S8 Digital photos of the VGs-Li₂S/LiPON/Pre-Li cell (sealed with CR2025-type coin cell) connected with a small thermometer



Fig. S9 The fitted Nyquist plots of the VGs-Li₂S/LiPON/Pre-Li cell after 1000 cycles



Fig. S10 The corresponding relationship between Z_{Re} and $\omega^{-1/2}$ in the low frequency region of after (a) 200, (b) 500 and (c) 1000 cycles

The Li⁺ diffusion coefficient has been calculated from plots in the low frequency region using the following equation:

$$D = R^2 T^2 / 2A^2 F^4 C^2 \sigma_W^2$$
 (S1)

Where R is the gas constant, T is the absolute temperature, A is the area of the cathode thin film, F is the Faraday constant, C is the concentration of Li^+ , and σ_W is the Warburg impedance coefficient, which can be obtained from the slope of the real part of resistance (Z_{Re}) and the inverse square root of angular frequency ($\omega^{-1/2}$).



Fig. S11 EDS elemental mapping of S, C, and P at the VGs-Li₂S/LiPON interface before cycling



Fig. S12 Voltage profiles of the VGs-Li₂S/LiPON/Evp-Li cell at the first three cycles under a current density of $1.12 \ \mu A \ cm^{-2}$



Fig. S13 FIB-SEM image of VGs-Li₂S/LiPON/Evp-Li cell after cycling