

SUPPLEMENTARY INFORMATION

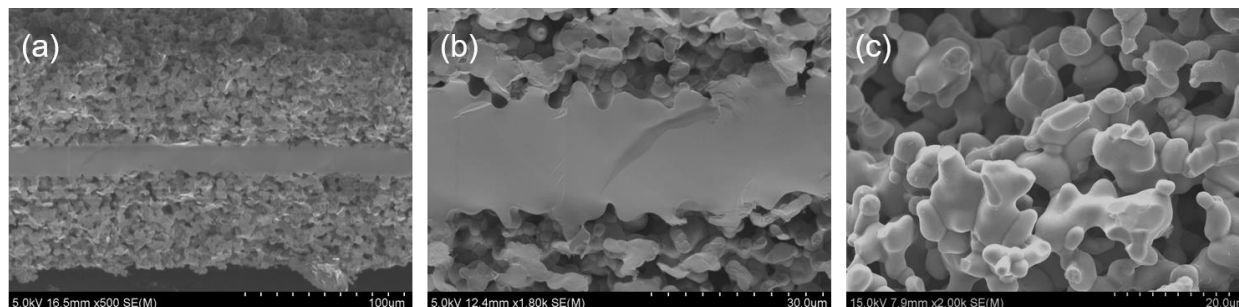


Figure S1. Cross-sectional SEM image of trilayer garnet. The trilayer garnet is composed of 15 μm thick dense garnet layer sandwiched by two 50 μm thick porous layers (a). The dense layer has no pinholes, which prevents lithium dendrite and lithium polysulfide from physically going through (b). The porous layer is composed of interconnected garnet grains which ensures ionic conductivity in the framework and has a high porosity for electrode infiltration (c).

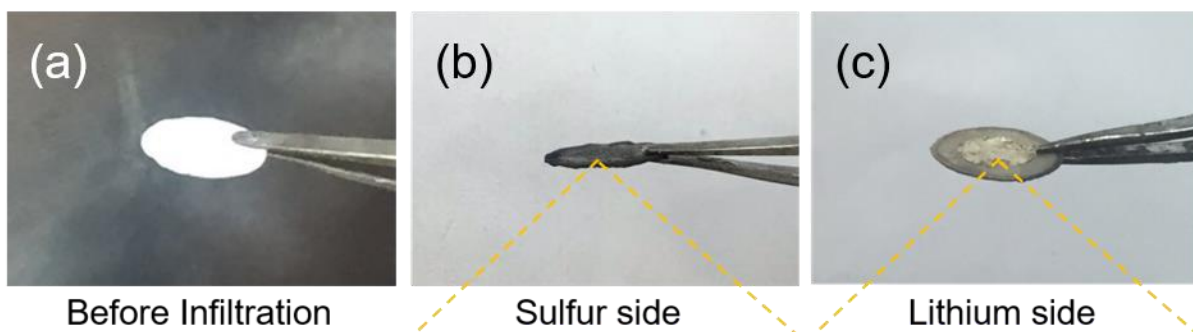


Figure S2. Digital photos of coin cell sized trilayer garnet before and after electrode infiltration. The pristine trilayer is a spherical ceramic plate with white color (a). After electrode infiltration, the cathode is filled with sulfur compound with CNT, which shows a black/grey color (b). The anode is filled with lithium metal which shows shiny metallic color (c).

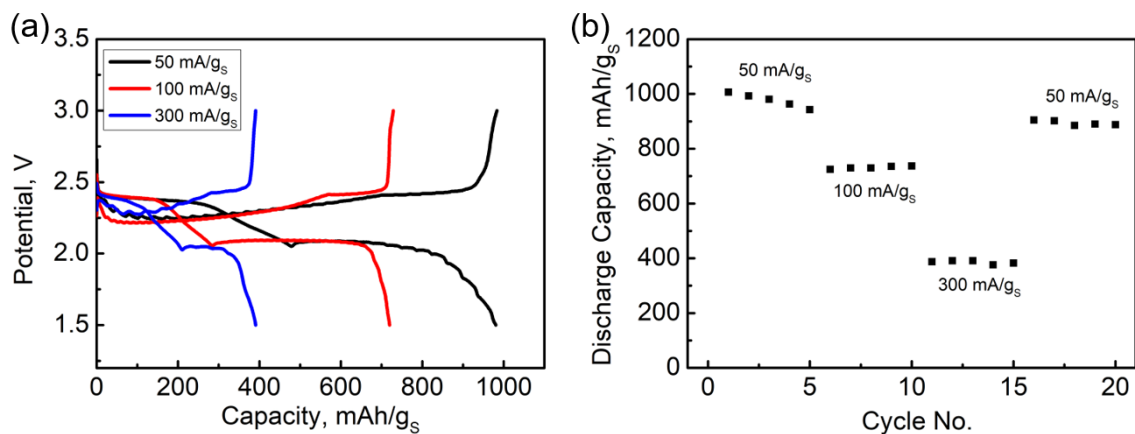


Figure S3. Rate performance of the all-in-one Li-S battery. (a) Voltage profiles of the trilayer Li-S cell with current densities of 50, 100 and 300 mA/g_s. (b) Cycling profile of the trilayer Li-S cell under different current densities. The cell shows stable cycling under elevated current densities and the capacity is recovered once the current density is reduced.

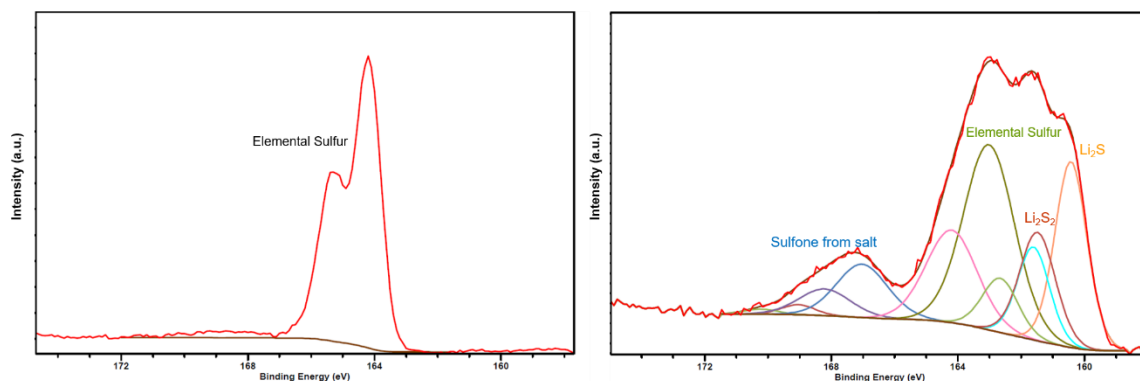


Figure S4. XPS spectrum of sulfur cathode in the (left) pre-cycling charged state and (right) post-cycling discharged state. In the charged state, only elemental sulfur is present, as expected in a fully delithiated sulfur cathode. Fully lithiated Li₂S₂ is present in the discharged state. Li₂S and elemental sulfur are still present, though, indicating incomplete sulfur utilization. The sulfone peaks are associated with the liquid electrolyte added for good interfacial contact.

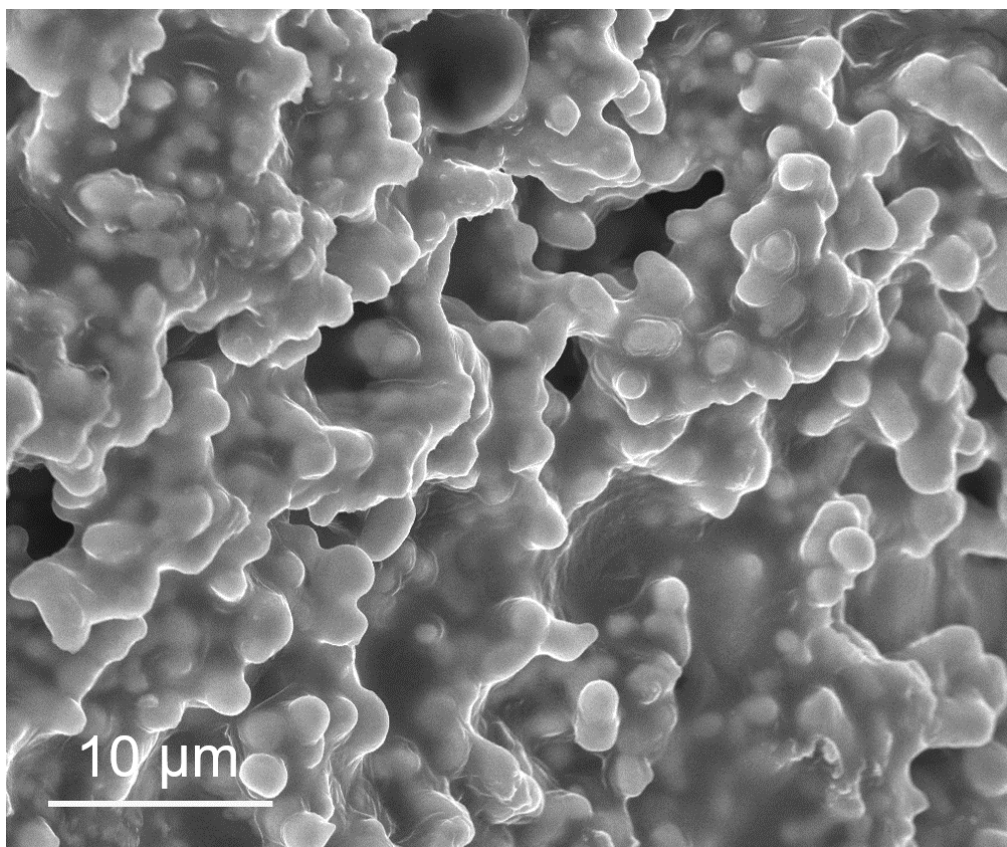


Figure S5. Post mortem SEM image of Li metal anode after cycling. The remaining lithium metal forms a thick coating on the garnet surface, maintaining the mixed electron/ion conductivity.

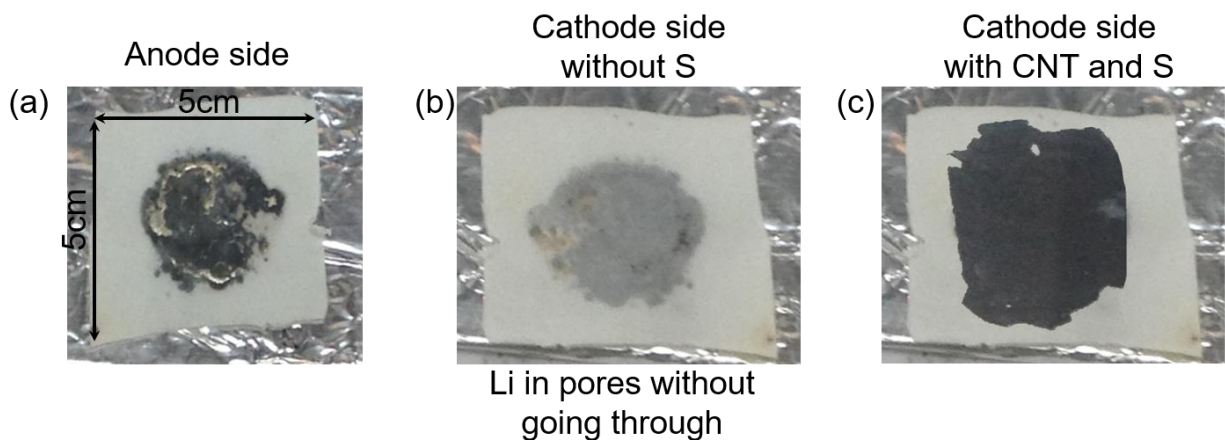


Figure S6. Scale up of the trilayer garnet after electrode infiltration. The lithium metal is infiltrated into anode pores and shows a metallic color on the surface of the trilayer garnet (a). Before sulfur infiltration, due to the translucency of thin ceramic, a dark shade can be observed from the cathode side due to the infiltration of lithium, indicating the pores are occupied by lithium metal (b). After sulfur infiltration, the cathode is filled with S/CNT cathode and show a black color from the surface (c).

Demonstrated

	Material	Volume ratio	Volume (cm ³)	Density (g/cm ³)	Mass (mg)	Specific Capacity (mAh/g)	Capacity (mAh)
Cathode 70 μ m	Garnet	30%	0.002	4.97	10.4	0	0.0
	Sulfur	39%	0.003	1.96	5.4	1,169	6.3
	Carbon	10%	0.001	1.7	1.2	0	0.0
	Empty	21%	0.001	0	0.0	0	0.0
Electrolyte 30 μ m	Garnet	100%	0.003	4.97	14.9	0	0.0
Anode 70 μ m	Garnet	30%	0.002	4.97	10.4	0	0.0
	Lithium	70%	0.005	0.54	2.6	3,861	10.2
	Empty	0%	0.000	0	0.0	0	0.0
Excess Lithium 31 μ m	Lithium	100%	0.003	0.54	1.6	3,861	6.4
Totals:			0.020		46.6		6.3
						Energy Density	272 Wh/kg
							633 Wh/L

Projected Performance

	Material	Volume ratio	Volume (cm ³)	Density (g/cm ³)	Mass (mg)	Specific Capacity (mAh/g)	Capacity (mAh)
Cathode 70 μ m	Garnet	30%	0.002	4.97	10.4	0	0.0
	Sulfur	39%	0.003	1.96	5.4	1,169	6.3
	Carbon	10%	0.001	1.7	1.2	0	0.0
	Empty	21%	0.001	0	0.0	0	0.0
Electrolyte 10 μ m	Garnet	100%	0.001	4.97	5.0	0	0.0
Anode 43 μ m	Garnet	30%	0.001	4.97	6.4	0	0.0
	Lithium	70%	0.003	0.54	1.6	3,861	6.3
	Empty	0%	0.000	0	0.0	0	0.0
Excess Lithium 3 μ m	Lithium	100%	0.000	0.54	0.2	3,861	0.6
Totals:			0.013		30.1		6.3
						Energy Density	421 Wh/kg
							1,007 Wh/L

Table S1. Calculation of specific energy and energy density for demonstrated cell, structurally optimized cell, and a structurally optimized cell achieving theoretical sulfur capacity. All calculations are on a per cm² basis. (**Demonstrated Cell**) The integrated capacity curve (from 0 to 1,169 mAh/g_{sulfur}) revealed that the demonstrated cell produced 2.37 Wh/g_{sulfur}. This energy and the masses of components from the table were used in the specific energy calculation for the “Demonstrated” cell. Specific energy and energy density are 272 Wh/kg_{cell} and 633 Wh/L_{cell}, respectively. (**Projected cell performance**) The “Projected” cell uses the demonstrated sulfur capacity (1,169 mAh/g_{sulfur}) to capacity match the electrodes by adjusting the anode layer thickness. Along with a minimization of excess lithium (5% excess for contact with current collector) and a more optimized 10 μ m dense layer, the mass of the optimized cell is calculated.

Because the sulfur loading is not changed, the same total energy is used as the demonstrated cell.

Specific energy and energy density are 421 Wh/kg_{cell} and 1,007 Wh/L_{cell}, respectively.