Electronic Supplementary Information

DNA-directed amphiphilic self-assembly as a chemifunctional/multiscale-structuring strategy for high-performance Li-S batteries

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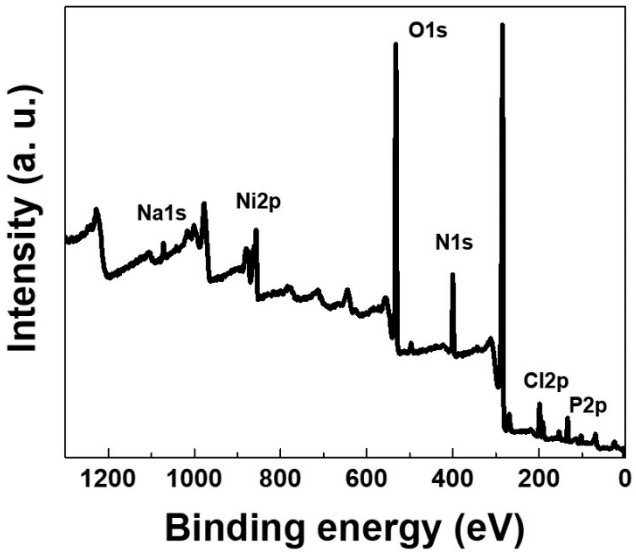

Atom	C	O	N	P	Na	Ni	Cl
%	65.6	18.4	7.3	2.5	0.5	1.3	3.0

Fig. S1. Survey scan XPS spectrum and summary of the atomic percent of the DNA/SWCNT hydrogel.

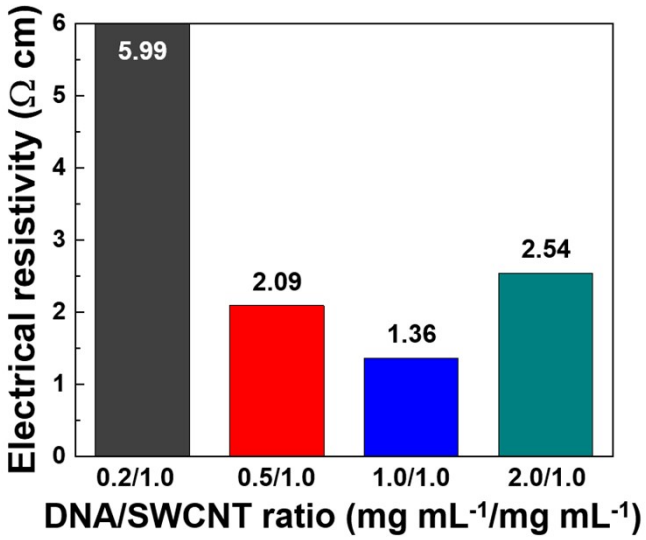

Fig. S2. Electrical resistivity of DNA/SWCNT films as a function of the DNA/SWCNT composition ratio.

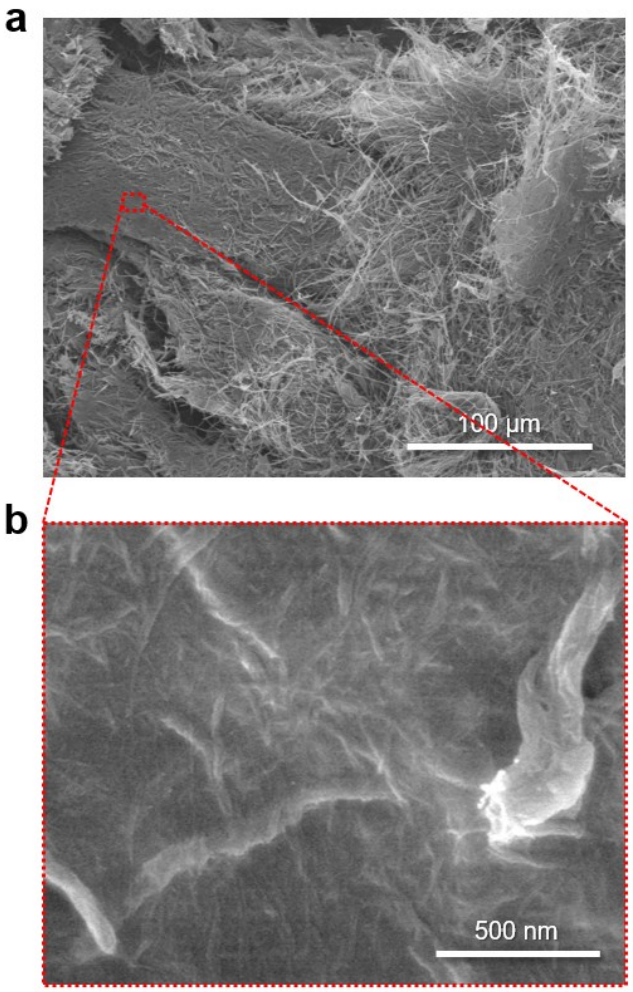

Fig. S3. SEM images of a control foam prepared using solely a DNA/SWCNT hydrogel (followed by freeze-drying) without the addition of Ni^{2+} ions. (a) Low and (b) High magnification.

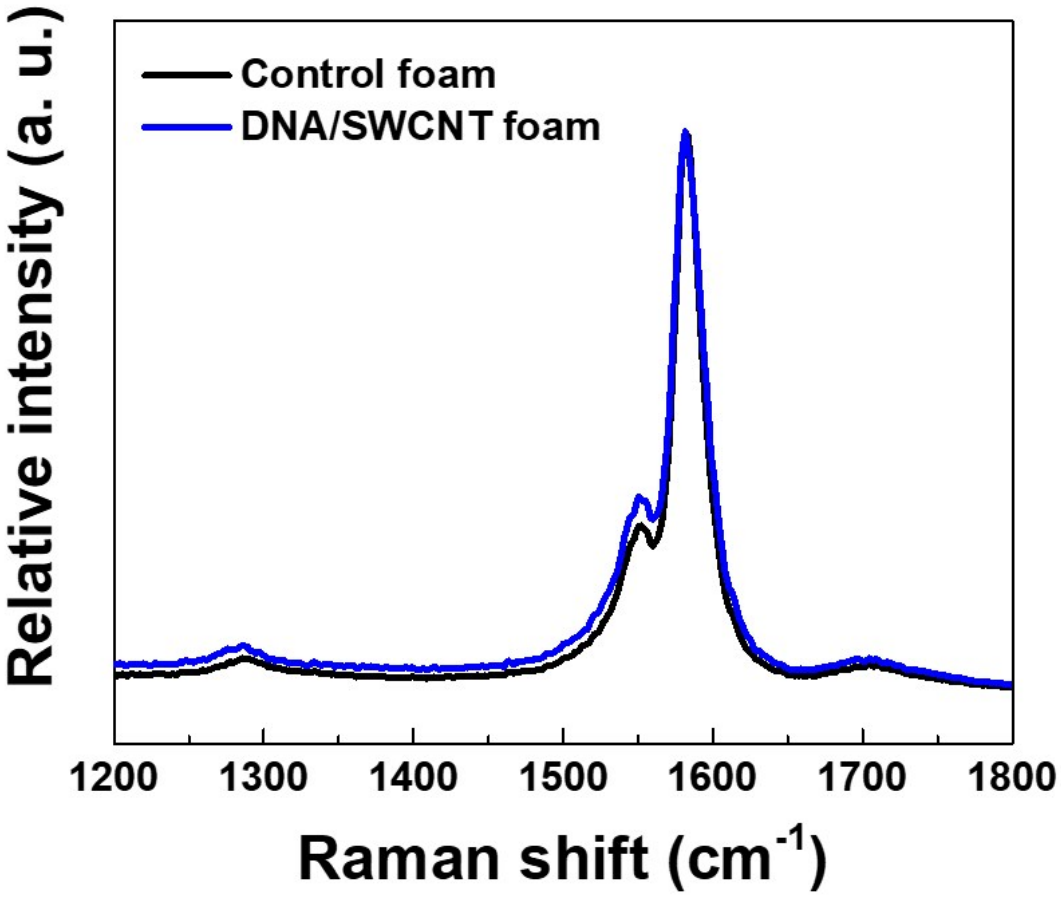

Fig. S4. Raman spectra showing the D and G band of the DNA/SWCNT foam (vs. control foam). No significant difference in the D/G ratio was observed between the DNA/SWCNT and control foams.

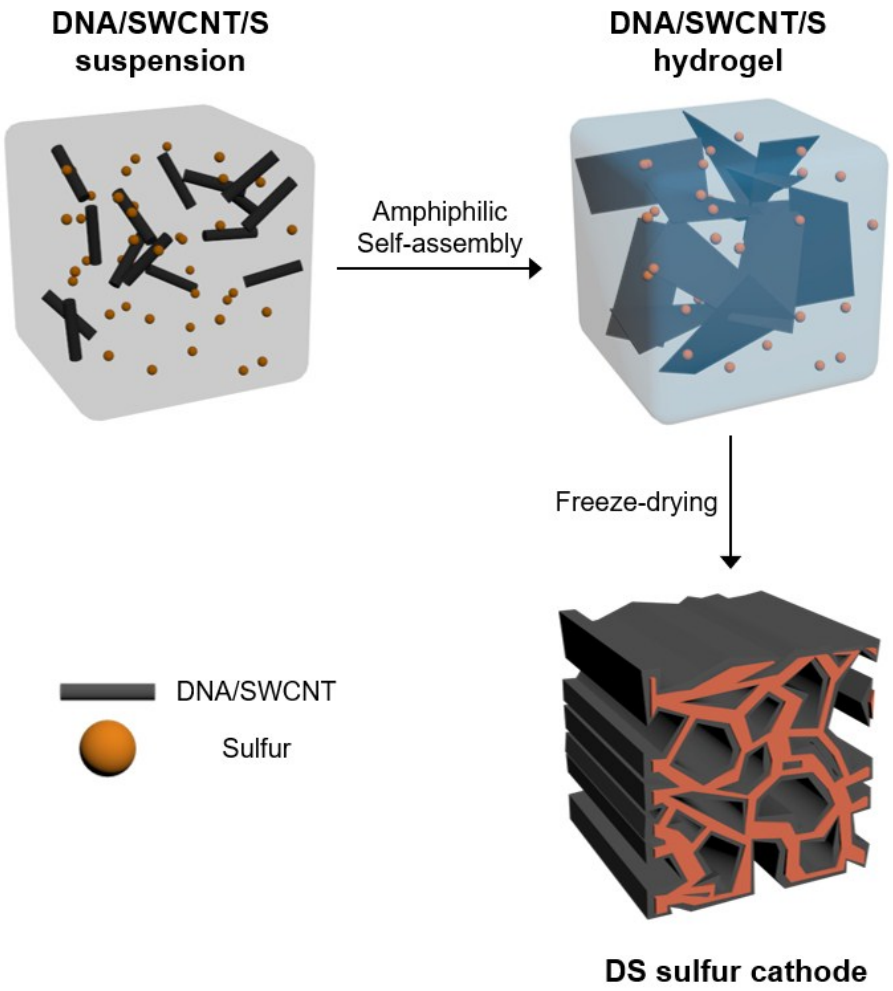

Fig. S5. Schematic representation depicting stepwise fabrication process of the DS sulfur cathode through the ASA process.

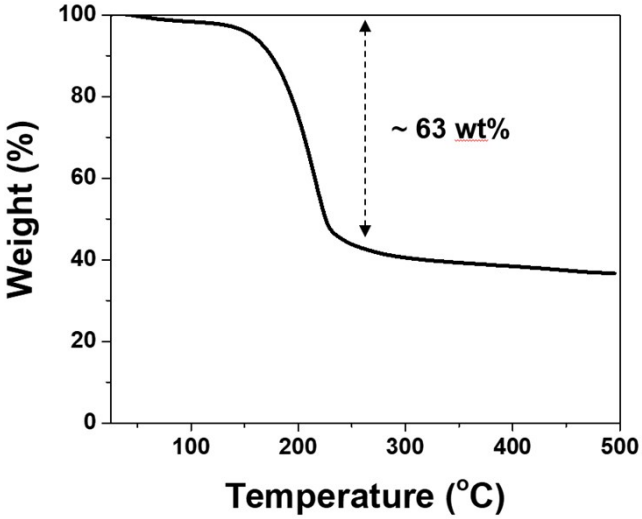

Fig. S6. TGA profile of the DS sulfur cathode. From this analysis, the sulfur content was quantitatively estimated.

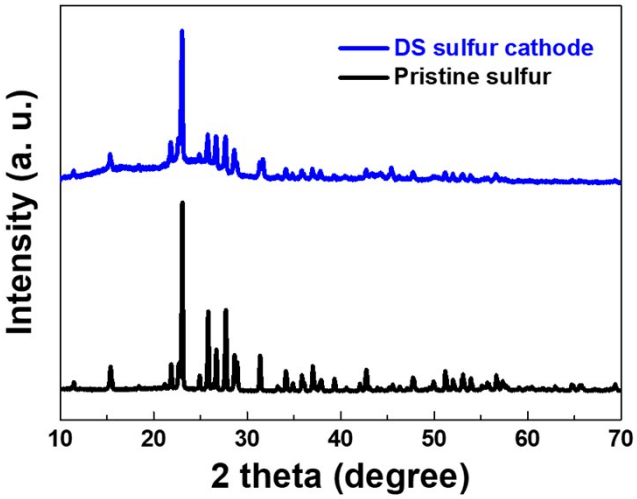

Fig. S7. XRD patterns of the DS sulfur cathode (vs. pristine sulfur).

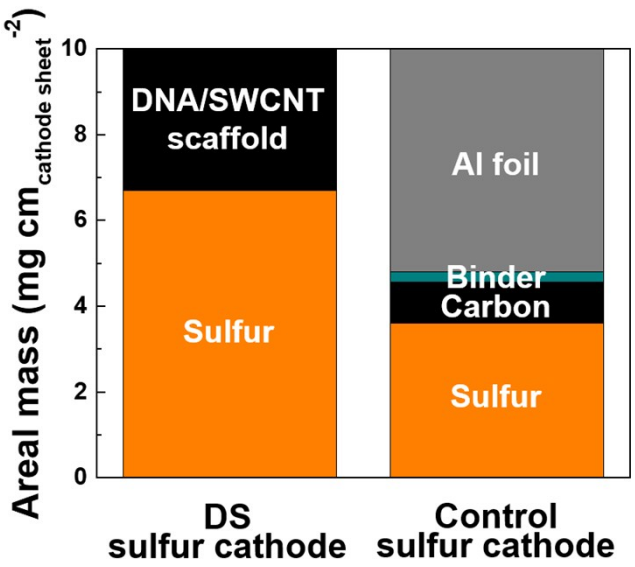

Fig. S8. Analysis of areal mass (= mg $cm_{cathode\ sheet}^{-2}$) of cathode components: DS sulfur cathode vs. control sulfur cathode.

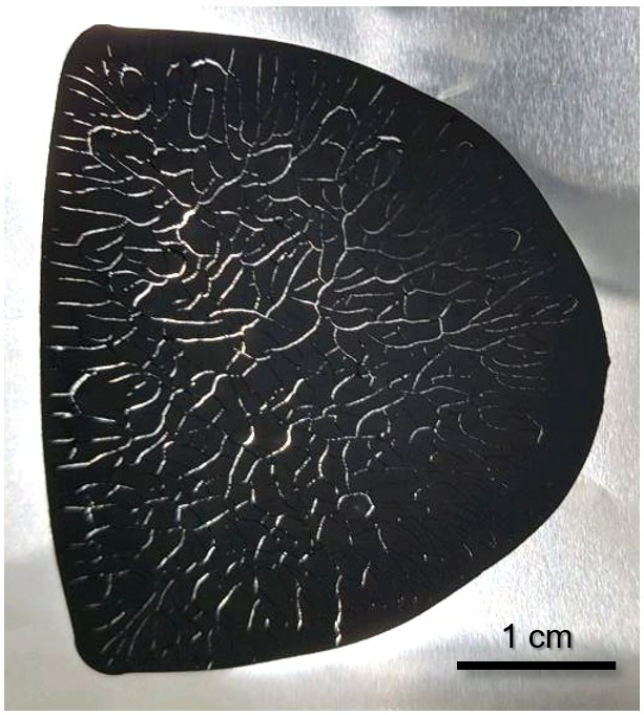

Fig. S9. Photo image of the control sulfur cathode (areal cathode weight ~ 12 mg cm^{-2}) showing the poor structural integrity.

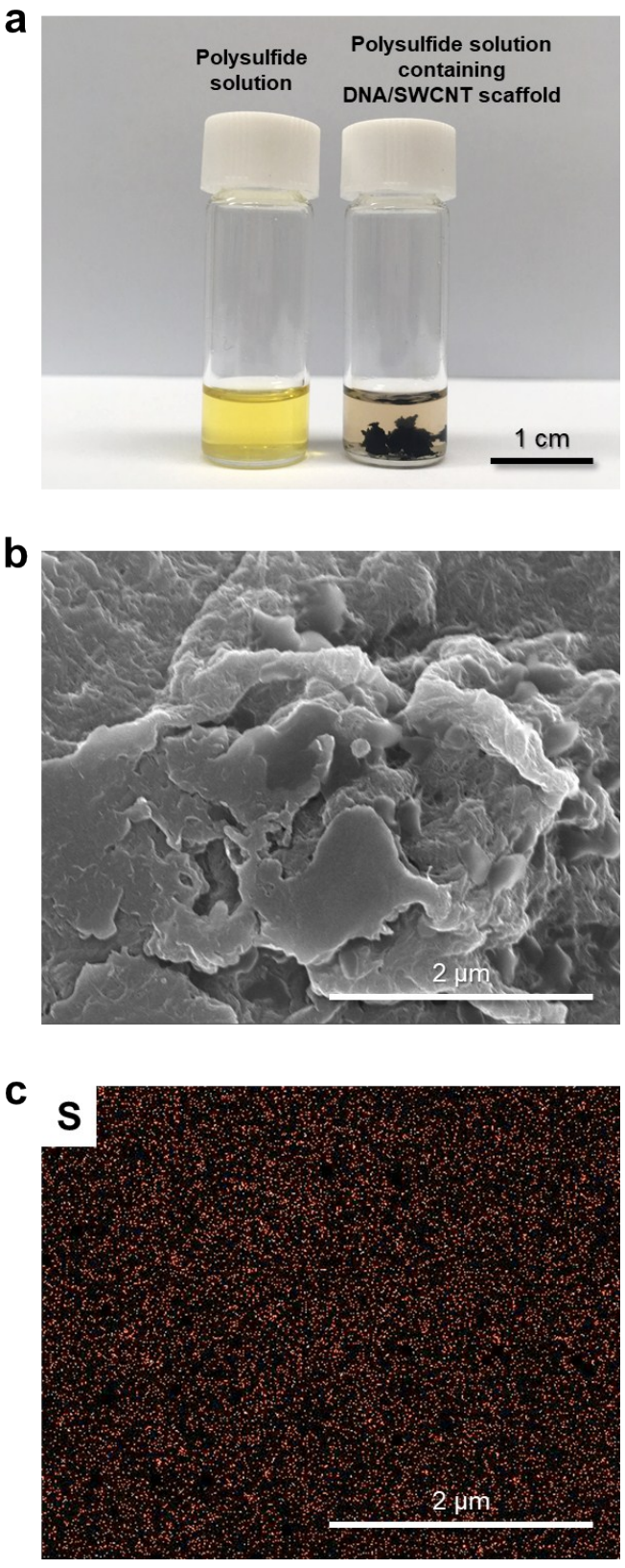

Fig. S10. (a) Photo, (b) SEM and (c) EDS (sulfur elements) images showing the polysulfide capturing capability of the DNA/SWCNT foam. The DNA/SWCNT foam (as a scaffold of the DS sulfur cathode) was dipped into a polysulfide solution (20 mM Li_2S_6 in DOL/DME = 1/1 (v/v)), and the color change of the solution was monitored after 24 h.

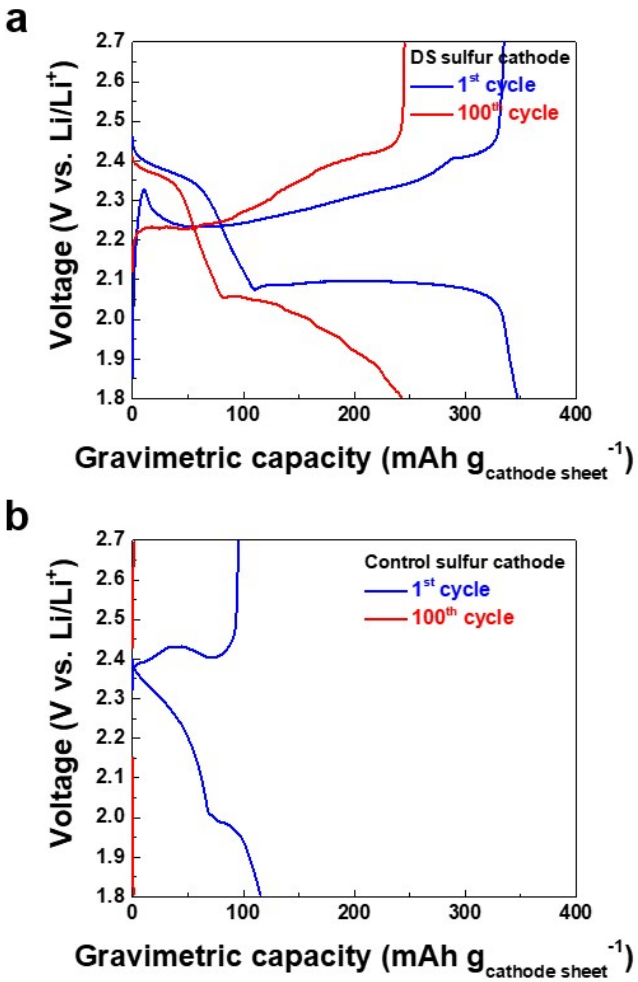

Fig. S11. Charge/discharge profiles (capacities are expressed as mAh $g_{cathode\ sheet}^{-1}$) of Li-S cells at 1st and 100th cycle. The cells were cycled at charge/discharge current densities of 0.1 C/0.1 C under voltage range of 1.8 – 2.7 V. The areal cathode weight was fixed at 10 mg $cm_{cathode\ sheet}^{-2}$. (a) DS sulfur cathode. (b) Control sulfur cathode.

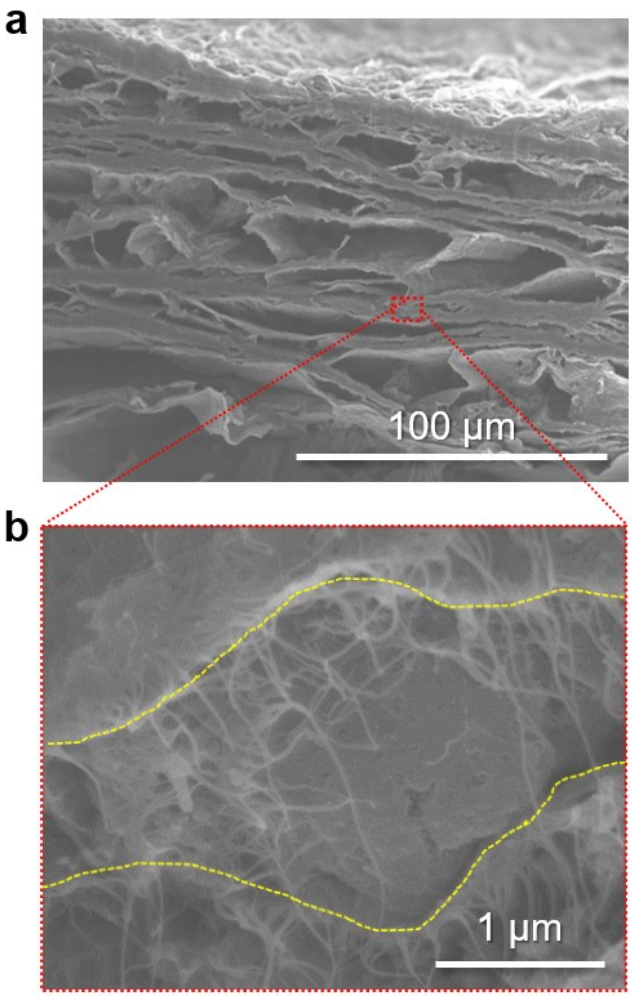

Fig. S12. Cross-sectional SEM image of the DS sulfur cathode after the cycle test.

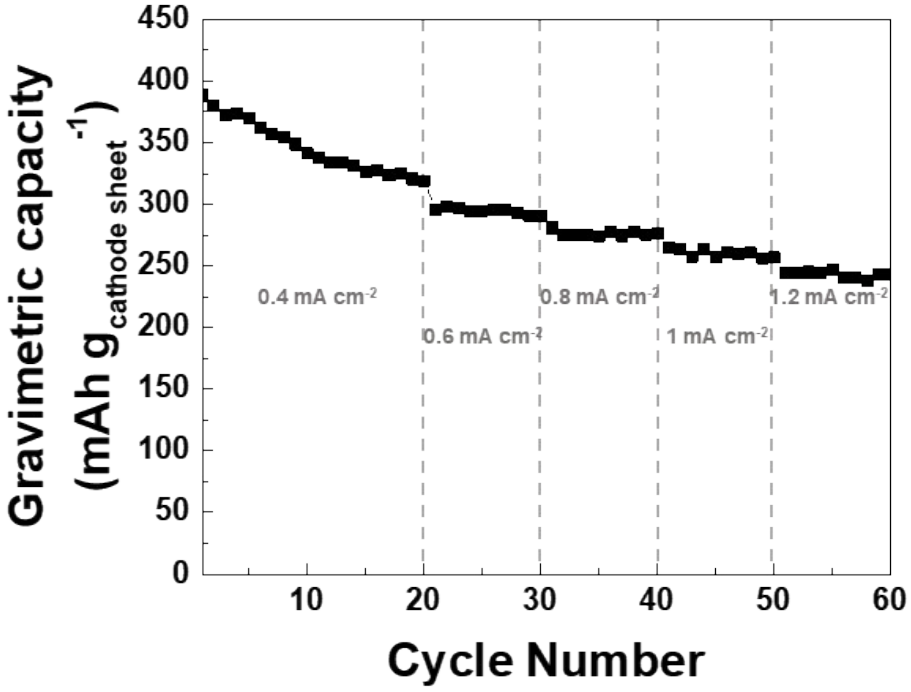

Fig. S13. Discharge capabilities of DS sulfur cathode as a function of discharge current density (varying from 0.4 to 1.2 mA cm^{-2}).

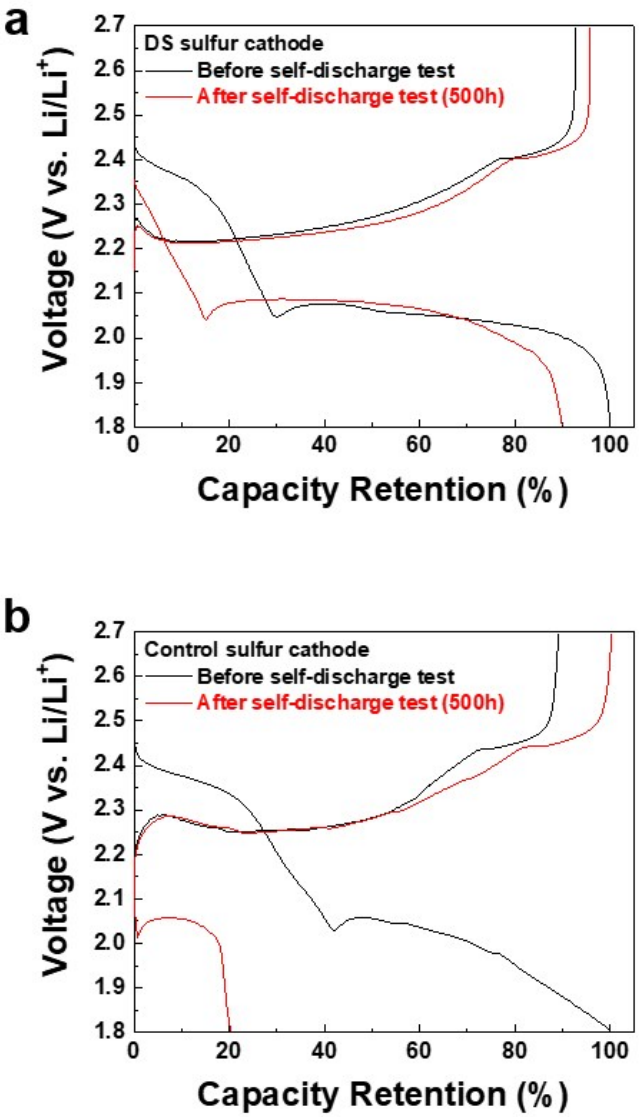

Fig. S14. Charge/discharge profiles of Li-S cells before/after the OCV test (elapsed time = 500 h). (a) DS sulfur cathode. (b) Control sulfur cathode.

Table S1. Comparison of assembly dimension: ASA process (this study) vs. previously reported DNA-mediated assembly techniques.

Approach		Assembly dimension		Reference
		x μm, y μm, z μm	μm^3	
2D assembly	Ribbon	2, 8, 2×10^{-3}	3×10^{-2}	*Nature*, 1998, **394**, 539-544.
	Lattice	3×10^{-1}, 2×10^{-1}, 2×10^{-3}	2×10^{-4}	*Nature*, 2006, **440**, 297-302.
3D assembly	Box	4×10^{-2}, 4×10^{-2}, 4×10^{-2}	5×10^{-5}	*Nature*, 2009, **459**, 73-76.
	Tube	2×10^{-2}, 2×10^{-2}, 6×10	2×10^{-2}	*Science*, 2008, **321**, 824-826.
	Curvatures	4×10^{-2}, 4×10^{-2}, 7×10^{-2}	2×10^{-2}	*Science*, 2011, **332**, 342-346.
	Bricks	4×10^{-2}, 3×10^{-3}, 8×10^{-2}	9×10^{-6}	*Science*, 2012, **338**, 1177-1183.
	Molecular force spectrometer	6×10^{-2}, 1×10^{-2}, 6×10^{-2}	4×10^{-5}	*Nat. Nanotechnol.*, 2016, **11**, 47-52. *Sci. Adv.*, 2016, **2**, e160094.
	Bricks	2×10^{-1}, 1×10^{-1}, 2×10^{-1}	4×10^{-3}	*Nature*, 2017, **552**, 72-77.
	Framework	2×10^{-1}, 2×10^{-1}, 2×10^{-1}	1×10^{-2}	*Nature*, 2017, **552**, 78-83.
Supra-molecular assembly	Colloid	2, 2, 3×10^{-3}	4×10^{-3}	*Nat. Mater.*, 2009, **8**, 519-525.
	Hydrogel	7×10^{3}, 7×10^{3}, 3×10^{3}	1×10^{11}	*Nat. Mater.*, 2006, **5**, 797-801.
		9×10^{3}, 6×10^{3}, 5×10	3×10^{8}	*Science*, 2017, **357**, 1126-1130.
	Template	8×10, 8×10, 2×10^{2}	1×10^{6}	*Adv. Mater.*, 2008, **20**, 466-470.
		1×10, 1×10, 1×10	1×10^{3}	*Nat. Chem.*, 2017, **9**, 466-472.
	ASA	1D: 3×10^{-3}, 3×10^{-3}, 2×10^{-1}	2×10^{-7}	This work
		2D: 1×10^{2}, 8×10, 2	2×10^{4}	
		3D: 2×10^{4}, 2×10^{4}, 1×10^{4}	4×10^{12}	

Table S2. Analysis of cell impedance (shown in Fig. 2d) using the equivalent circuit model.

SWCNT network	$R_{bundle}(\Omega)$	$R_{junction}(\Omega)$	$R_{overall}(\Omega)$
Control foam	317	1013	1330
DNA/SWCNT foam	65	203	268

Table S3. Summery of the gravimetric capacities (based on the weight of cathode sheet and sulfur powders) of DS sulfur cathodes.

Loading level ($mg\ cm_{cathode\ sheet}^{-2}$)	Capacity based on cathode sheet ($mAh\ g_{cahode\ sheet}^{-1}$)	Capacity based on sulfur ($mAh\ g_{Sulfur}^{-1}$)
5.2	389.4	618.1
6.4	389.2	617.8
10.0	347.0	550.8
12.6	333.3	529.1
14.0	326.3	517.9