Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting information

Light-weight functional layer on separator as polysulfide immobilizers to enhance cycling stability for lithium-sulfur batteries

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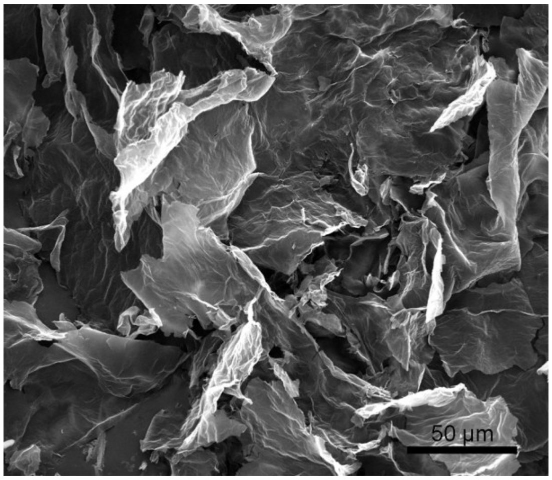

Figure S1 SEM image of the rGO sample.

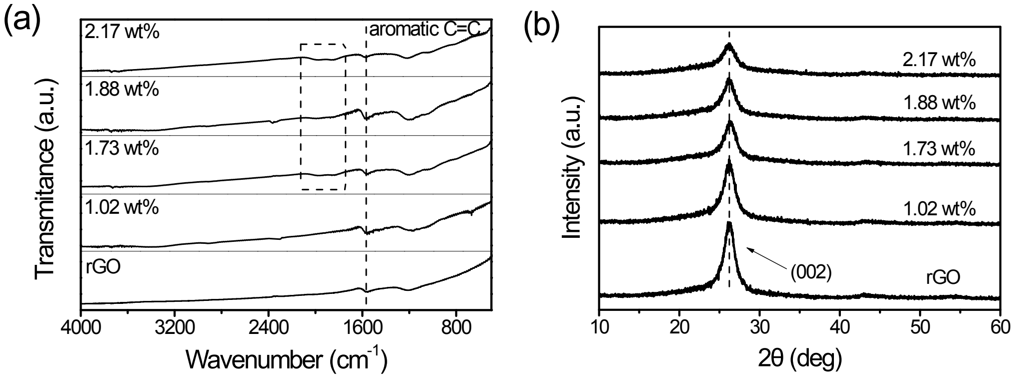

Figure S2 (a) FT-IR and (b) XRD patterns of the boron-functionalized rGO with different contents of boron.

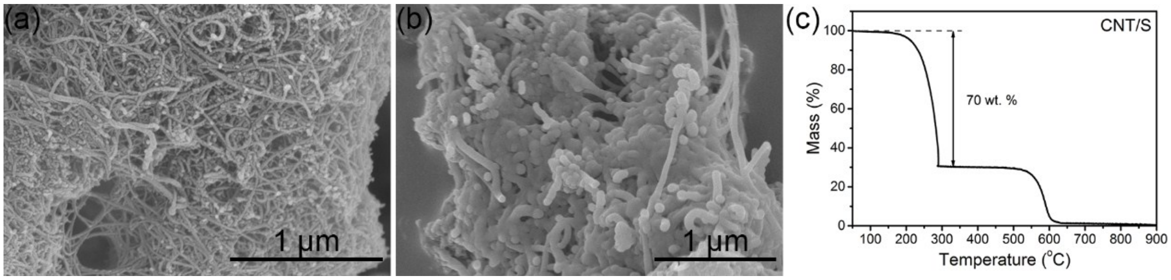

Figure S3 SEM images of (a) CNTs and (b) CNT/S composite. (b) TGA curves of the CNT/S composite, and the sulfur content is 70 wt. %.

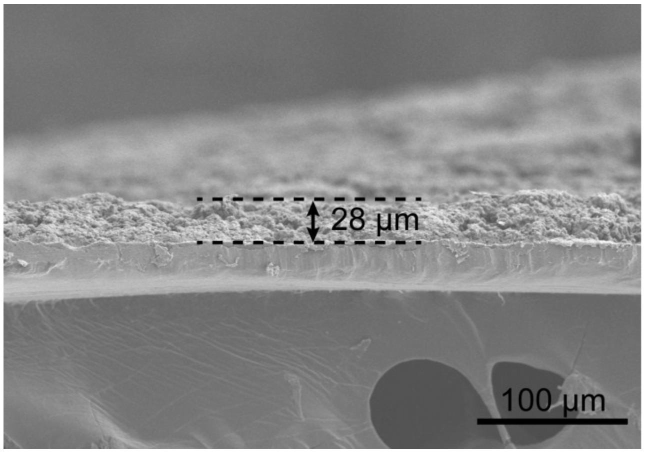

Figure S4 Cross section of the thin cathode. The sulfur content of composite for thin cathode is 70 wt. % and the sulfur loading for thin cathode is 1.45-1.56 mg/cm^2. As shown in Figure S4, the thickness of the thin cathode is 28 µm.

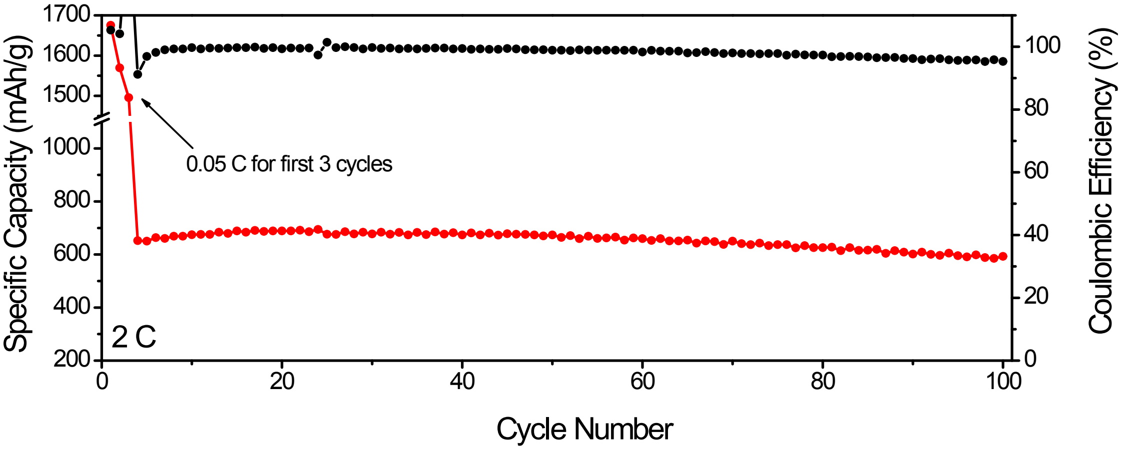

Figure S5 Cycle performance and corresponding coulombic efficiency of the cell with B-rGO coated separator at the charge-discharge current rate of 2 C.

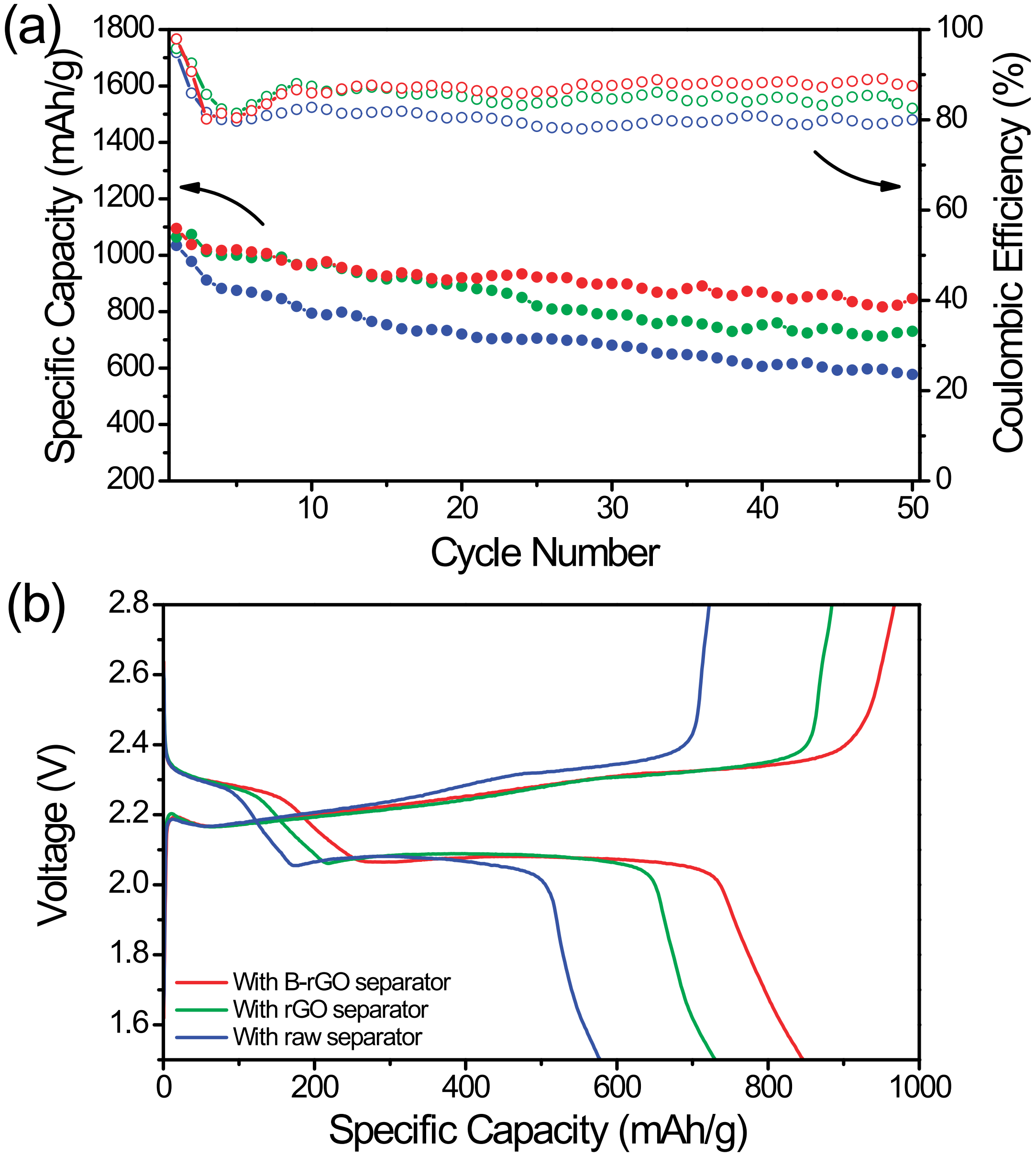

Figure S6 (a) Cycle performance and corresponding coulombic efficiency of the cells with different separators. (b) Discharge/charge voltage profiles in the 50th cycle of the Li-S cell. The cells were all cycled at a current rate of 0.2 C, and the voltage range is 1.5–2.8 V. The electrolyte used was 1.0 mol/L bis(trifluoromethane) sulfonimide lithium (LiTFSI) in dimethoxyethane (DME) and 1,3-dioxolane (DOL) solvents (1:1 v/v). The amount of electrolyte added into the cell was 50 μL for all the cathodes.

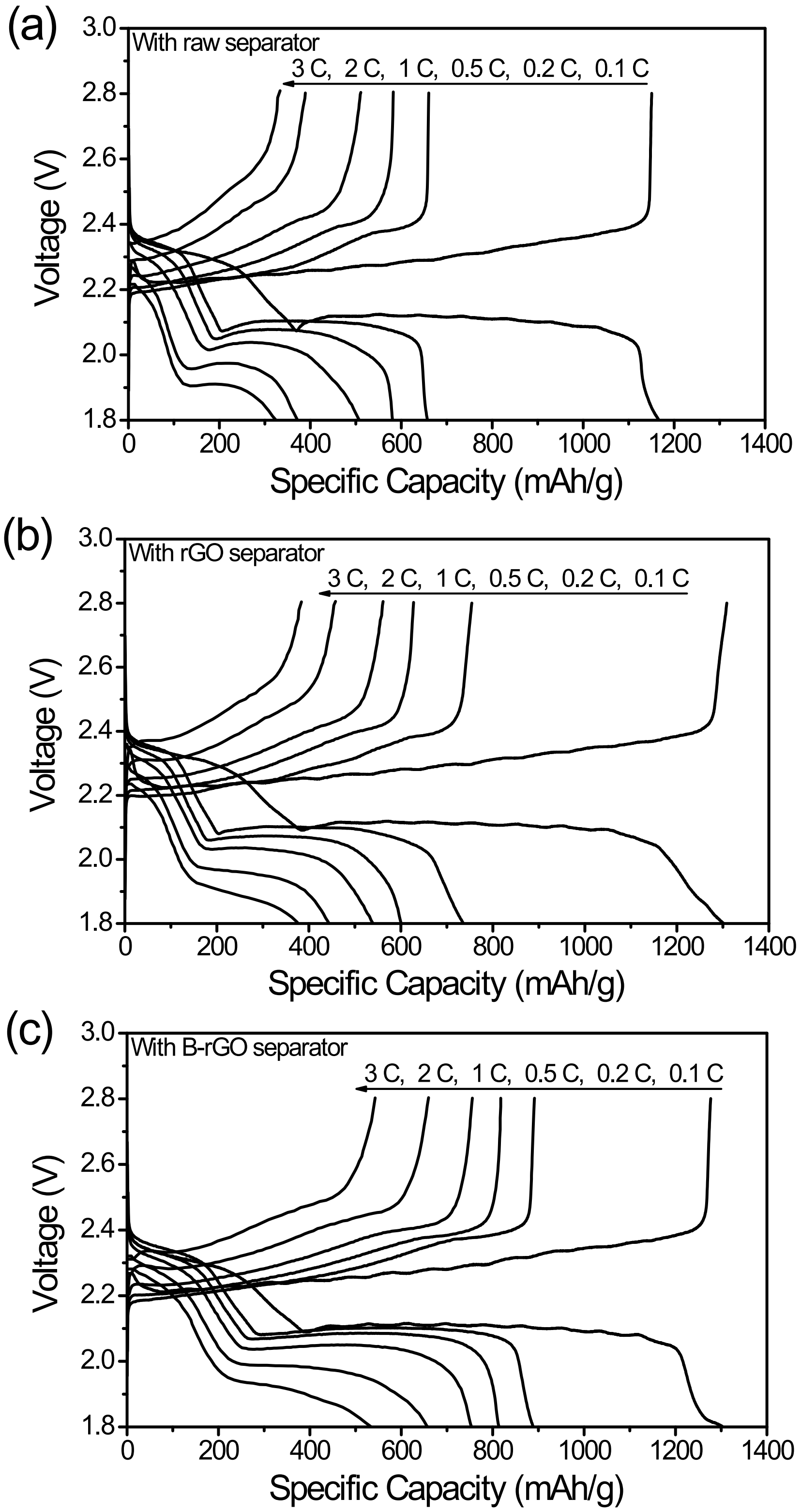

Figure S7 Discharge/charge voltage profiles of the cells with raw and rGO/B-rGO coated separators under different discharge/charge current rates.

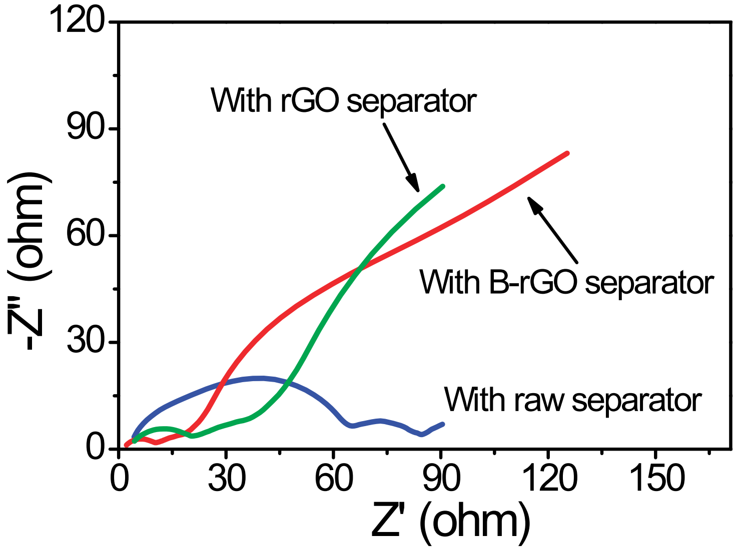

Figure S8 AC impedance measurements of cells containing different separators after 50 cycles at 0.1 C

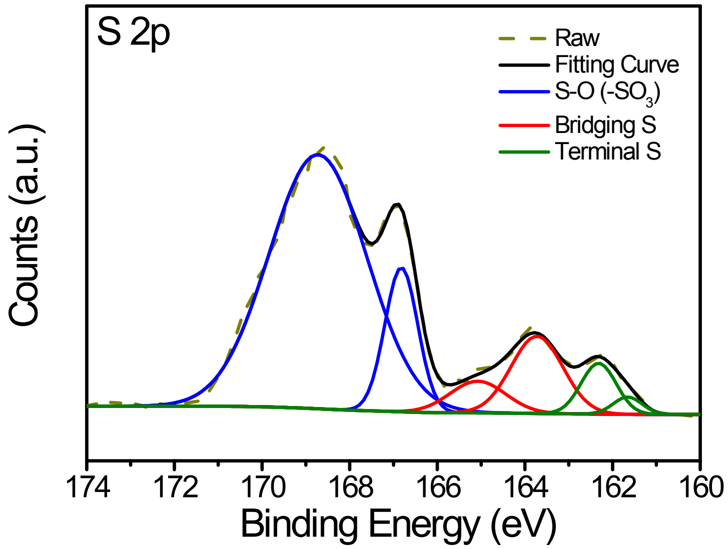

Figure S9 High-resolution XPS characterization of the S 2p region for the B-rGO coating layer on the separator after 100 cycles at 0.1 C.

It can be observed that three S 2p doublets centered at 162.3, 163.9, and 168.1 eV in the S 2p region. The S 2p doublet located at higher binding energy are assigned to sulfate compounds originating from the electrolyte salt.[1] The S 2p doublets positioned at lower binding energy is assigned to bridging and terminal sulfur species, indicating that long-chain polysulfide compounds exist within the B-rGO coating layer.

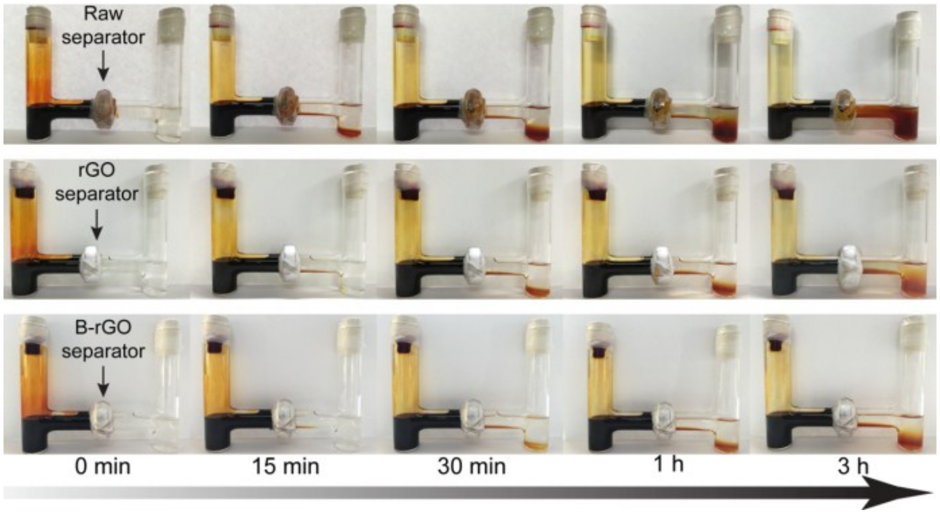

Figure S10 Photographs of polysulfides diffusion processes through the raw separator, rGO and B-rGO coated separator, which is tested by using a H-shaped cell.

The H-shaped cell separated by separator was used to test their ability in inhibiting polysulfides diffusion (Figure S7). 10 mL of the electrolyte with 0.5 M Li_2S_6 added and 10 mL blank electrolyte are added into the left and right sides of the cell, respectively. At the beginning, the right side of the cell is transparent, but it turns to yellow with the passing of time. With the raw separator, the color of the electrolyte changes to blood red thoroughly after 3 hours. Meanwhile, the rGO and B-rGO coated separators can effectively mitigate the polysulfides diffusion, and the role of B-rGO layer is more notable. These results provide support on better performance of the cells with B-rGO coated separator.

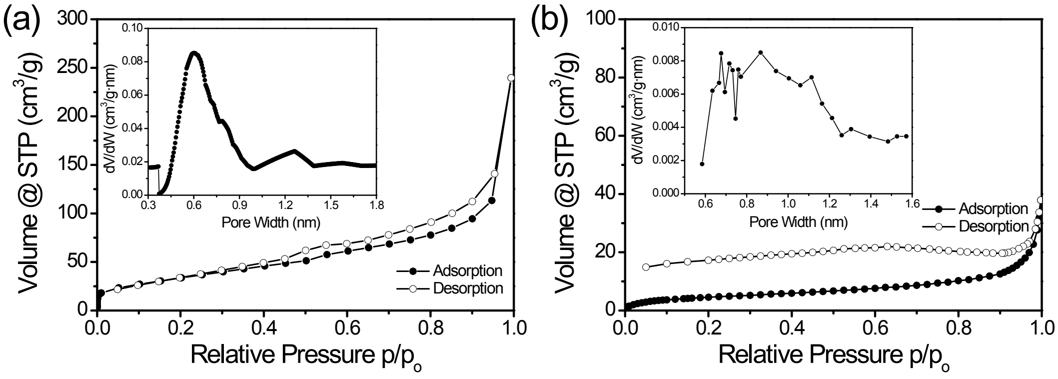

Figure S11 Nitrogen sorption isotherms and pore size distribution for (a) B-rGO and (b) rGO. The specific surface area (based on Brunauer–Emmett–Teller method) values of B-rGO and rGO are 127.819 and 17.6538 m²/g, respectively.

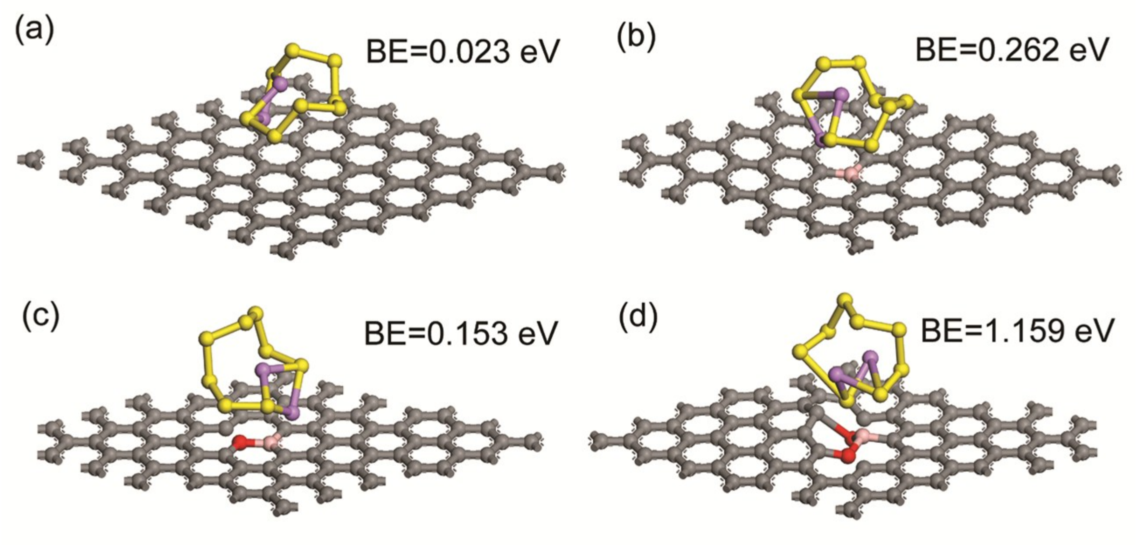

Figure S12 Optimized geometries of the Li_2S_8 molecule and (a) raw graphene, (b-d) three typical forms of the boron-functionalized graphene. The binding energy (BE) values are also listed. Gray, light magenta, red, purple, and yellow balls represent C, B, O, Li, and S atoms, respectively. Optimized geometries of the Li_2S_8 molecule adsorbed on the graphene surfaces are presented, and corresponding binding energy values are listed. The raw graphene shows the lowest binding energy of 0.023 eV with Li_2S_8, and the energy values increase to 0.262, 0.153, 1.159 eV for three typical forms of the boron-functionalized graphene, respectively.

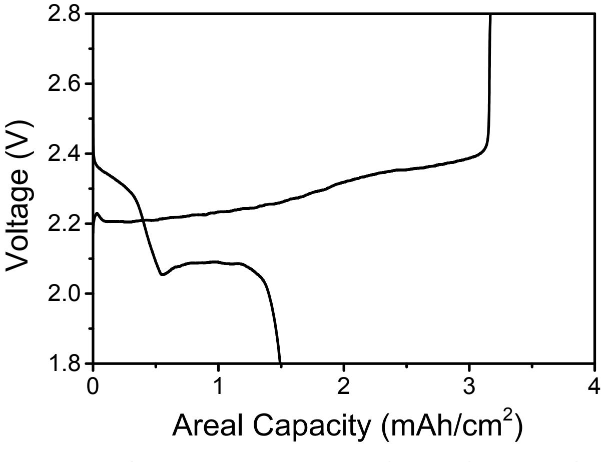

Figure S13 The 18th discharge/charge voltage profiles of the Li-S cell with thick cathode and raw separator under the discharge/charge current density of 0.389 mA/cm^2.

Table S1 Summary of electrochemical performances of representative modified separators for Li-S batteries.

Sample	Weight loading of sample on separator (mg/cm^2)	Cathode materials (sulfur content in composite * percentage of composite in cathode)	Areal sulfur loading (mg/cm^2)	Discharge rate	Initial discharge specific capacity (mAh/g)	Cycle	Capacity decay rate per cycle	Ref.
B-rGO coated Celgard 2032 separator	0.2-0.3	S/CNT (70% * 80%)	1.45-1.56	0.1 C	1227.8	300	0.1532%	This work
				2 C	652.2	100	0.0936%	
		S/MWCNT (90% * 70%)	4.60-4.67	0.389 mA/cm^2	4.7075 mAh/cm^2	35	0.5429%	
MesoC-Coated Separator Celgard 2500 separator	-	Sulfur–carbon composite (70% * 70%)	ca. 1.55	0.5 C	1216	500	0.081%	1
				1 C	1060		0.071%	
				2 C	856		0.062%	
super P coated Glassfiber membrane	-	Pure sulfur (70%)	0.7–1.0	0.2 C	1352	-	-	2
				1 and 2 C	-	200	721 and 607 mAh/g remained	
			2.13	0.2 C	1218	30	0.4%	
			3.37	0.2 C	867	30	0.2%	
Nafion coated Celgard 2400 separator	0.7	CNT-S (50% * -)	0.53	1 C	781	500	0.08%	3
	3.5				ca. 780		ca. 0.082%	
super P coated Celgard separator	0.2	Pure sulfur (60%)	1.1–1.3	0.2 C	1400	200	0.2043%	4
F-rGO coated on glass microfiber (Whatman)	0.3-0.4	S/C (50% * 80%)	1.5-2	0.1 C	ca. 1240	100	ca. 0.57%	5
					ca. 880		ca. 0.27%	
					ca. 750		ca. 0.4%	
GO coated Celgard 2400 separator	0.12	Pure sulfur (63%)	1.0-1.5	0.1 C	920	100	0.23%	6
	0.24			0.2 C	1040	50	ca. 0.55%	
N/P dual doped graphene coated Celgard 2300 separator	ca.1.0±0.1	Pure sulfur (70%)	-	1 C	1158.3	500	0.0898%	7
PEDOT:PSS coated Celgard 2500 separator	0.07	Sulfur-expanded graphene composite (80% * 80%)	0.9-1.1	0.25 C	985	1000	0.0364%	8

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