

## Supporting Information

### **A separator-based lithium polysulfides recirculator for high-loading and high-performance Li-S batteries**

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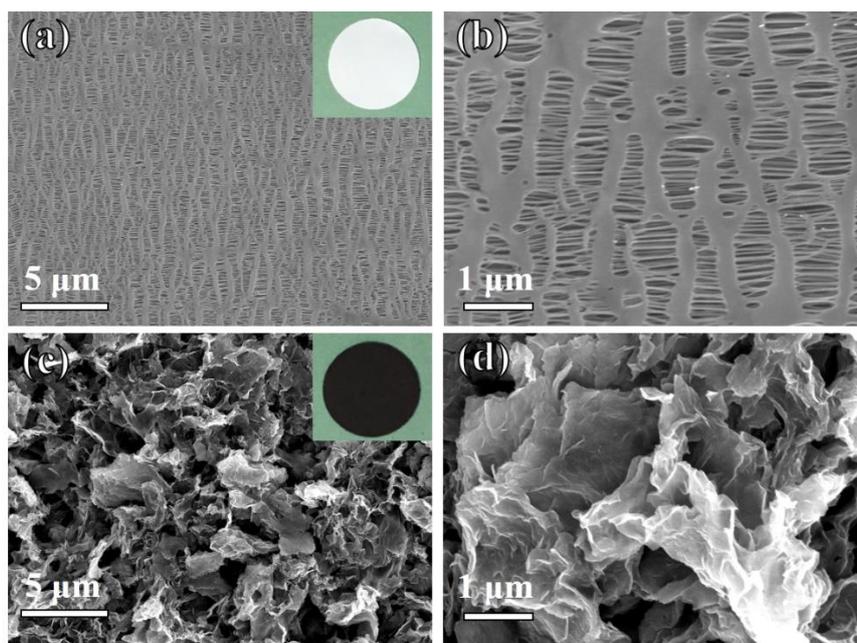
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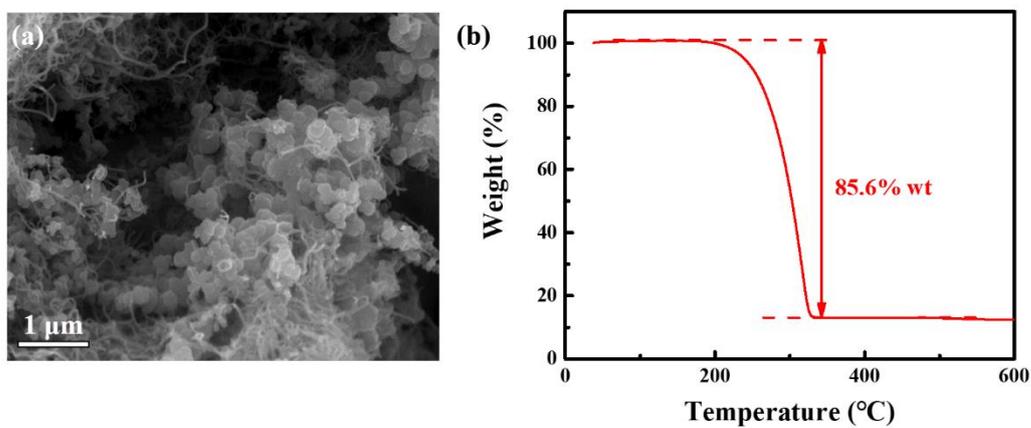
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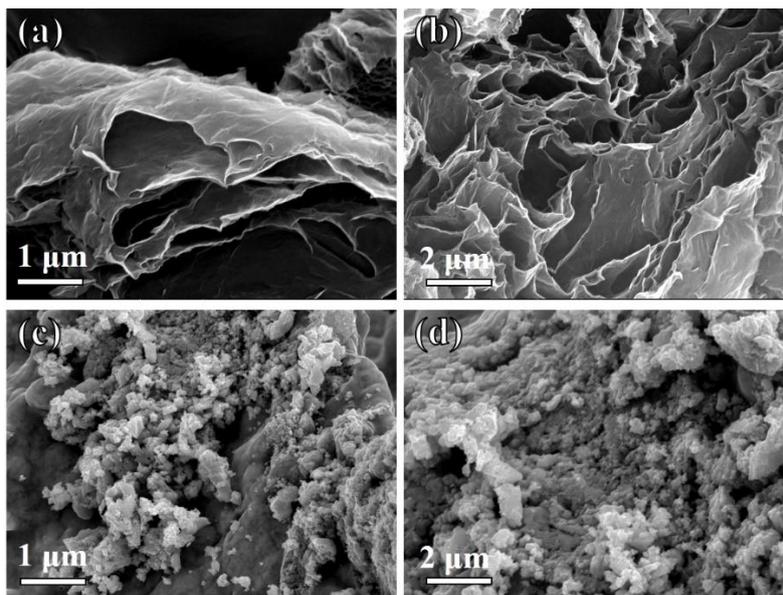
<sup>‡</sup> Chao Wang and Ming Li contributed equally to this work.



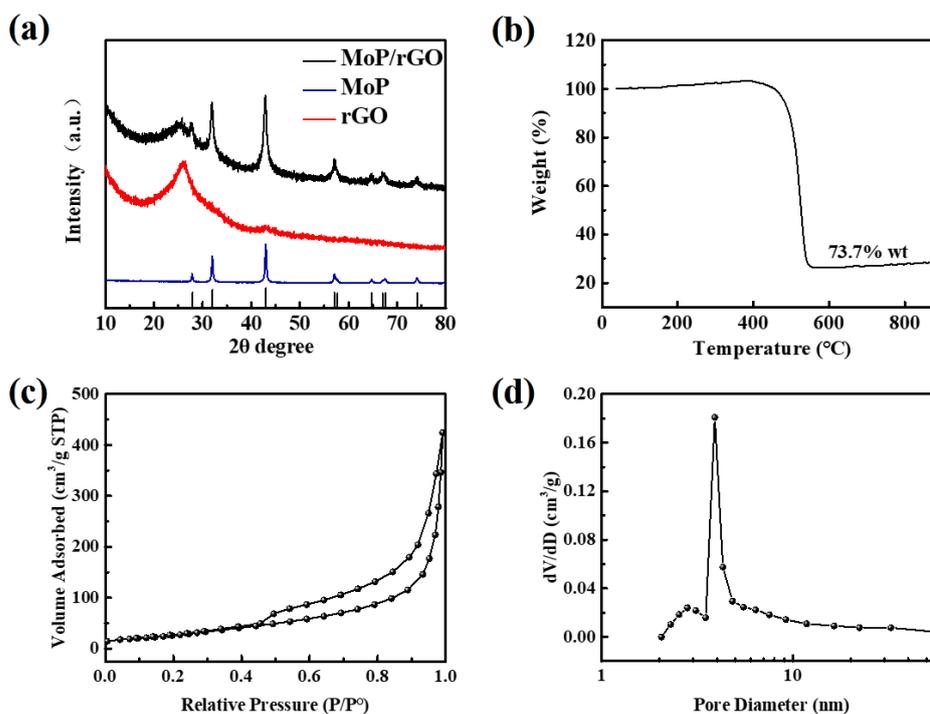
**Fig. S1** (a) (b) The digital photo and SEM images of Celgard separator. (c) (d) The digital photo and SEM images of MoP/rGO modified separator.



**Fig. S2** (a) The SEM image of S/C composites. (b) The TGA curve of S/C composites under Ar atmosphere.



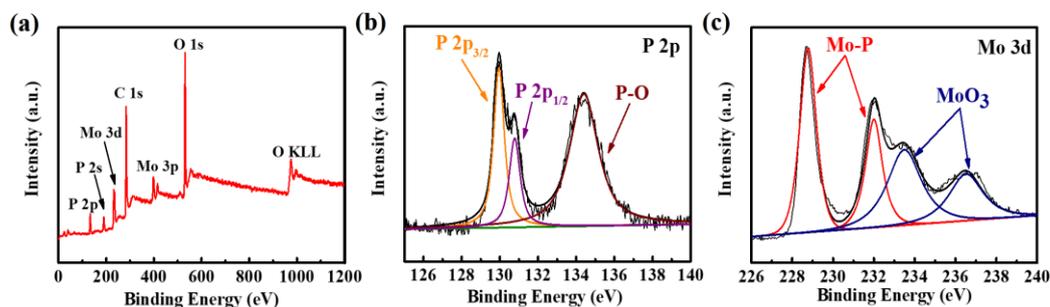
**Fig. S3** (a) (b) The SEM images of pure rGO. (c) (d) The SEM images of bulk MoP.



**Fig. S4** (a) XRD patterns of MoP/rGO, rGO and MoP. (b) TGA curve of MoP/rGO composites under air atmosphere (the final product is  $\text{MoO}_3$ ). (c)  $\text{N}_2$  adsorption-desorption isotherms of MoP/rGO composites. (d) The pore size distribution of the MoP/rGO composites.

To confirm the rGO content of the composites, the TGA measurement was made at 0~600 °C under air atmosphere. The final product of MoP/rGO composites was  $\text{MoO}_3$ . From the TGA results, the mass of  $\text{MoO}_3$  was about 26.3 % of the total mass.

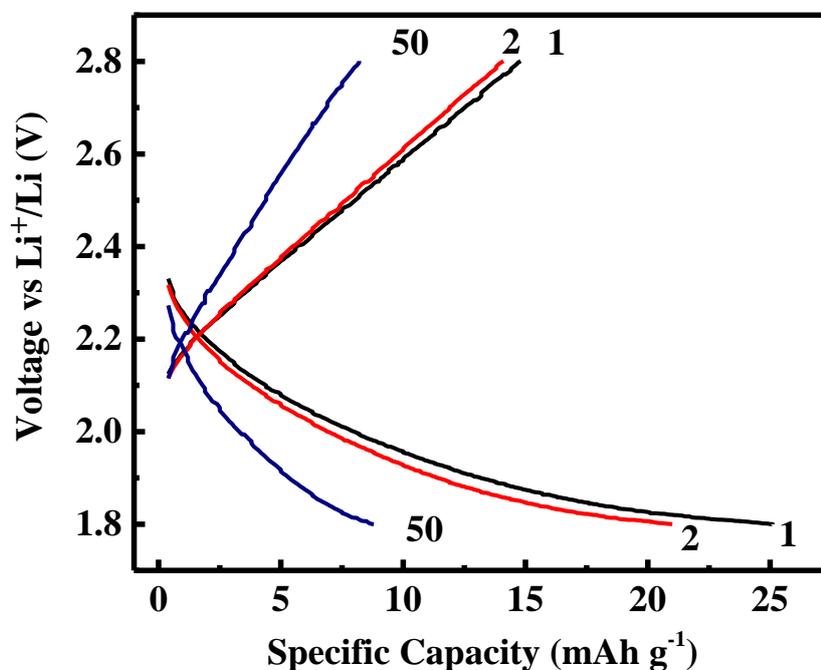
So the content of MoP in initial composites was  $26.3 \% * 127/144 = 23.2 \%$  (the relative molecular mass of MoP and MoO<sub>3</sub> are 127 and 144, respectively), which means the rGO content of the composites was about 76.8 %.



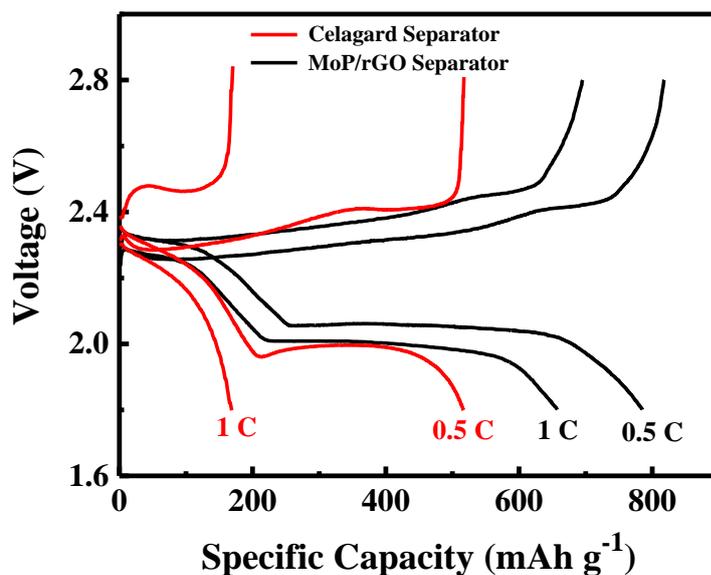
**Fig. S5** (a) wide-scan, (b) P 2p and (c) Mo 3d XPS spectra of MoP/rGO composites.

### Polysulfides Adsorption Experiment

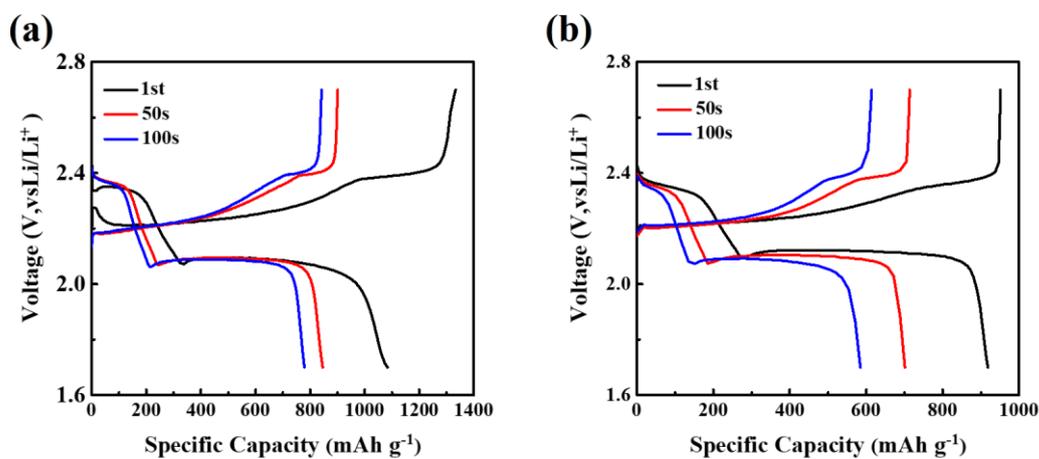
The Li<sub>2</sub>S<sub>6</sub> solution was prepared as follows. S<sub>8</sub> and Li<sub>2</sub>S were dissolved in 1, 3-dioxolane (DOL) and 1, 2-dimethoxyethane (DME) (1:1 by volume) with a molar ratio of 5:8. Then the mixture was stirred at 60 °C for 24 h in Argon-filled glovebox. The concentration of the obtained Li<sub>2</sub>S<sub>6</sub> solution was 1 mg mL<sup>-1</sup>. An equivalent amount (10 mg) of different polysulfide host materials (MoP/rGO, rGO and bulk MoP for comparison) were immersed into a 2 mL Li<sub>2</sub>S<sub>6</sub> solution and held for 2 h.



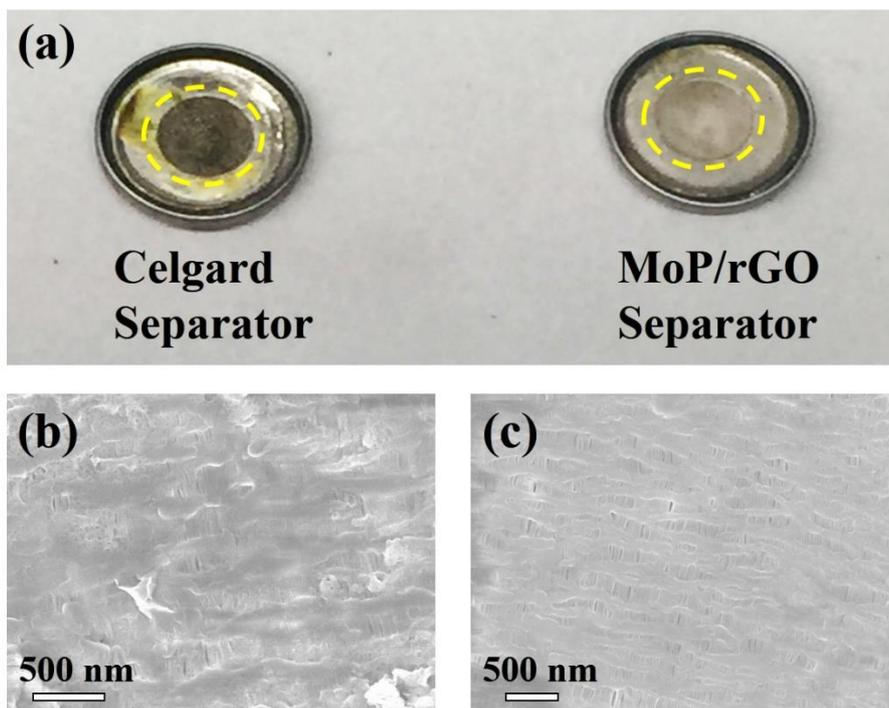
**Fig. S6** Different cycles of galvanostatic discharge-charge curves of MoP/rGO composite between 1.8 and 2.8 V at 0.5 C.



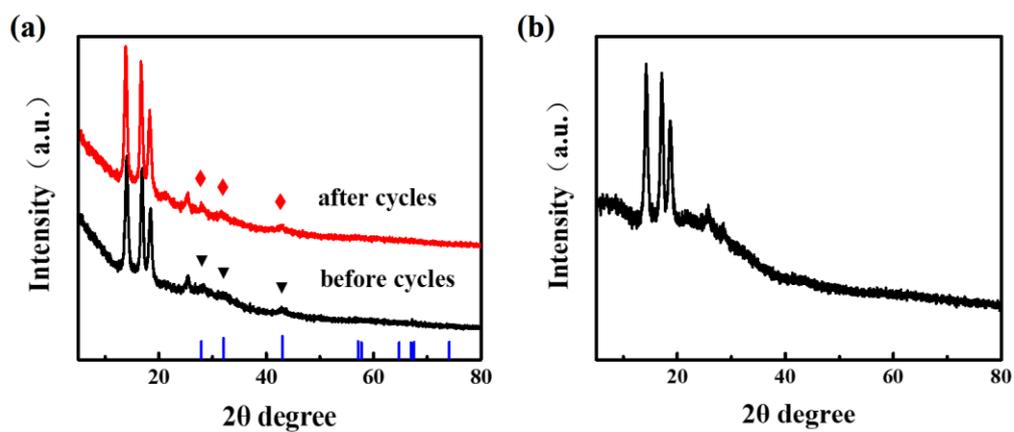
**Fig. S7** Galvanostatic discharge-charge profiles of cells with different separators at rate of 0.1C and 0.5C.



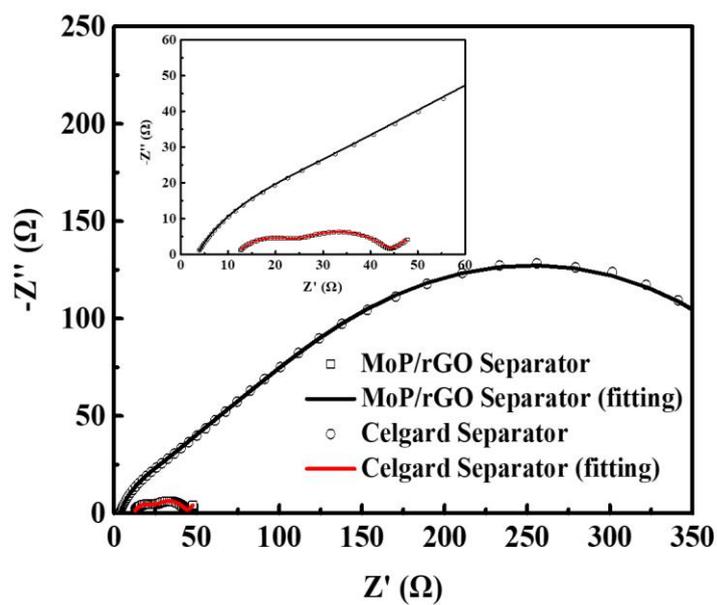
**Fig. S8** Galvanostatic discharge-charge profiles of pouch cells with (a) MoP/rGO modified separator, (b) Celgard separator separators at different cycles.



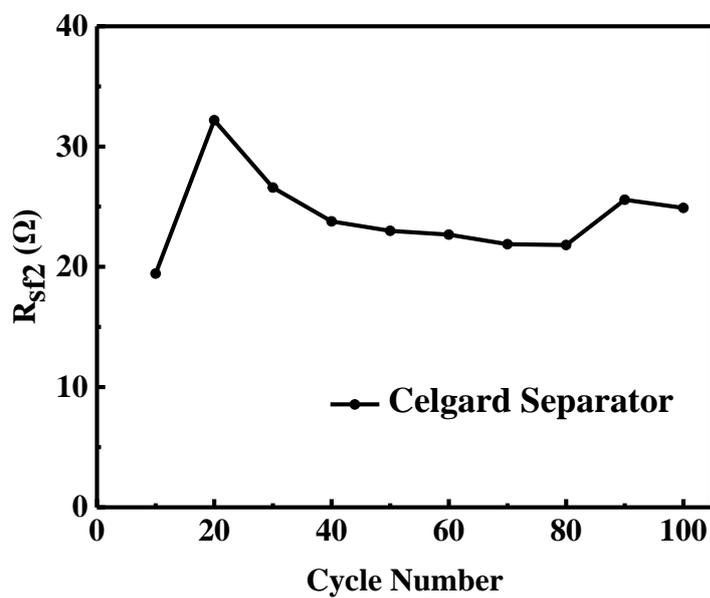
**Fig. S9** (a) Photograph of lithium anodes in cells with different separators after cycling. (b) (c) SEM images of the surface faced to the lithium anode of Celgard and MoP/rGO coated separator, respectively.



**Fig. S10** (a) XRD patterns of MoP/rGO modified separator before and after cycles; (b) XRD patterns of Celgard separator.



**Fig. S11** The electrochemical impedance spectra of cells with MoP/rGO coated separator and Celgard separator in full charge state after 100 cycles.



**Fig. S12** Interface resistance of  $R_{sf2}$  of cell with Celgard Separator in full charged states for 100 cycles.

**Table S1** Comparison of performance of Li-S batteries with different interlayers

Interlayers	Interlayers mass loading (mg cm <sup>-2</sup> )	Sulfur mass loading (mg cm <sup>-2</sup> )	Cathode (Sulfur content)	Electrochemical performance			
				Rate	Initial Capacity (mAh g <sup>-1</sup> )	Cycles	Capacity decay rate
MoP/rGO	0.35~0.45	3.6 ~ 4	CNT/S (77%)	0.1 C	1125	100	0.15%
				0.5 C	880	300	0.045%
Li <sub>4</sub> Ti <sub>5</sub> O <sub>12</sub> / graphene	0.346	1.0 ~ 1.2	CNT/S 60%	1 C	813	500	0.029%
Boron-rGO	0.2~0.3	1.5	CNT/S (56%)	0.1 C	1227	300	0.153%
TiO <sub>2</sub> /graphene	0.13	1.2	CNT/S (82%)	0.5 C	802	250	0.072%
TiO <sub>2</sub> /carbon	0.2	2	C/S (60%)	0.5 C	920	200	0.087%
MoS <sub>2</sub>	N/A	N/A	C/S (65%)	0.5 C	808	600	0.083%
MWCNTs/ NCQDs	0.15	1.3 ~ 1.5	C/S (60%)	0.5 C	1330	500	0.1%
Black P	0.4	1.5 ~ 2	C/S (66%)	0.5 C	930	100	0.14%
GO	0.12	1 ~ 1.5	CNT/S (63%)	0.1 C	920	100	0.23%

## Reference:

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