

Supplementary Materials

# Ultrahigh-capacity Lithium Metal Batteries Based on Multi-electron Redox Reaction of Organopolysulfides including Conductive Organic Moieties

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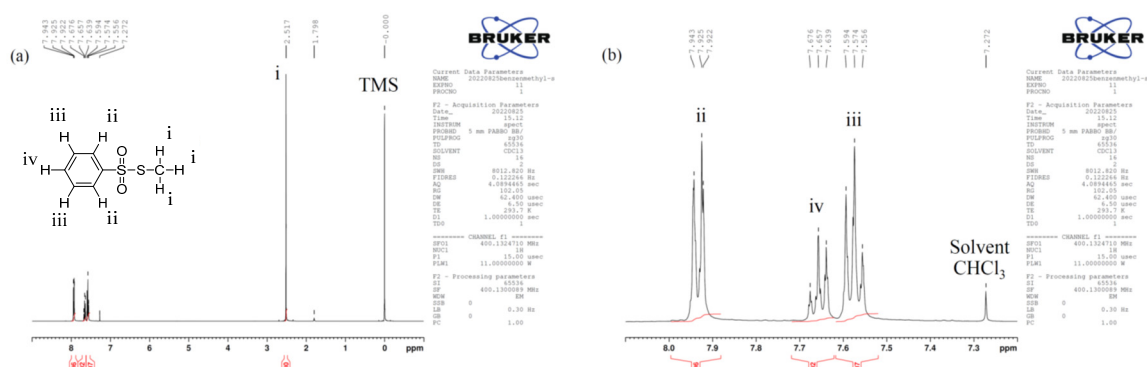


Figure S1. <sup>1</sup>H NMR charts of molecular 1, 0-8 ppm (a) and 7.2-8.0 ppm (b).

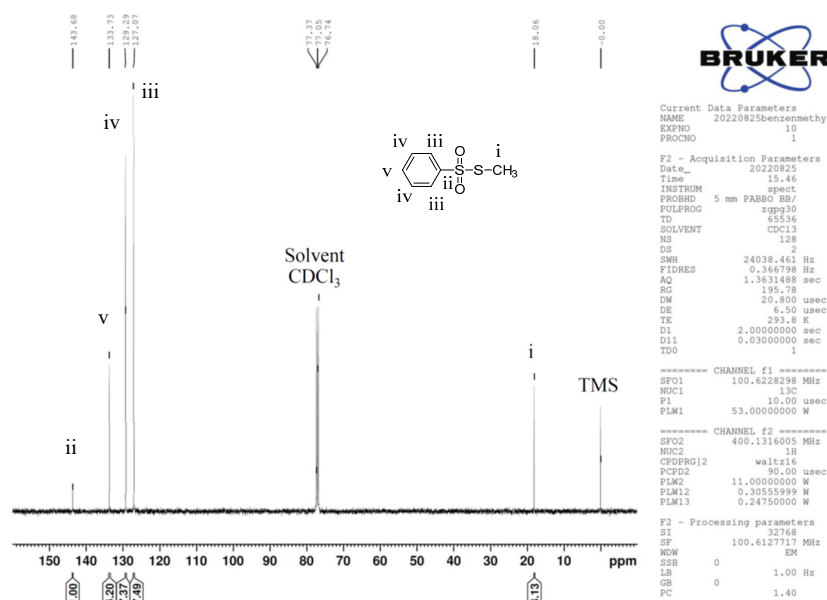
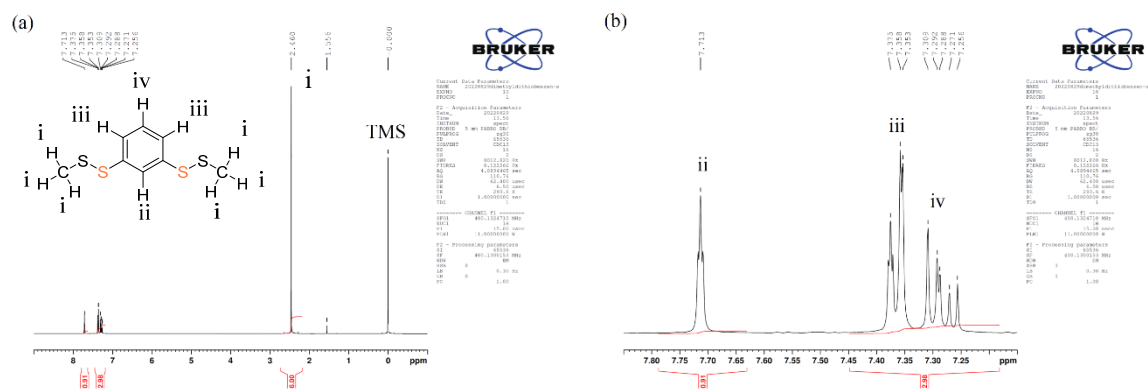
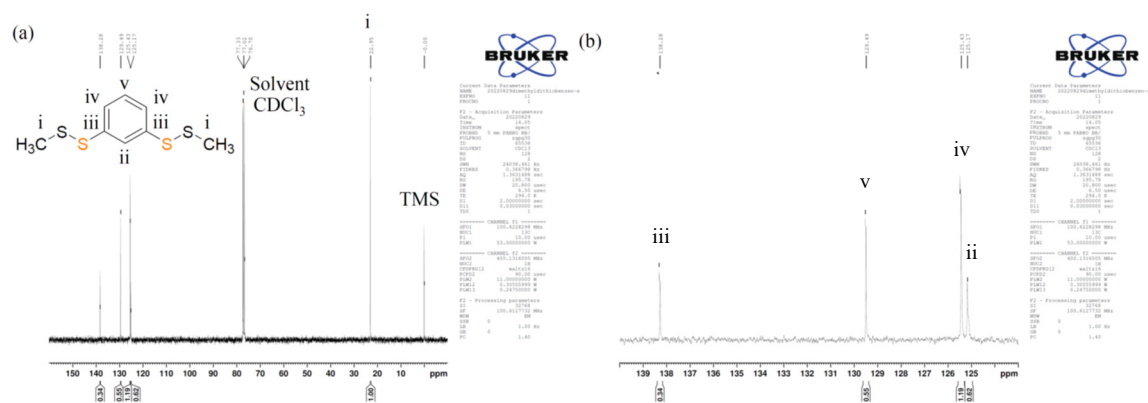
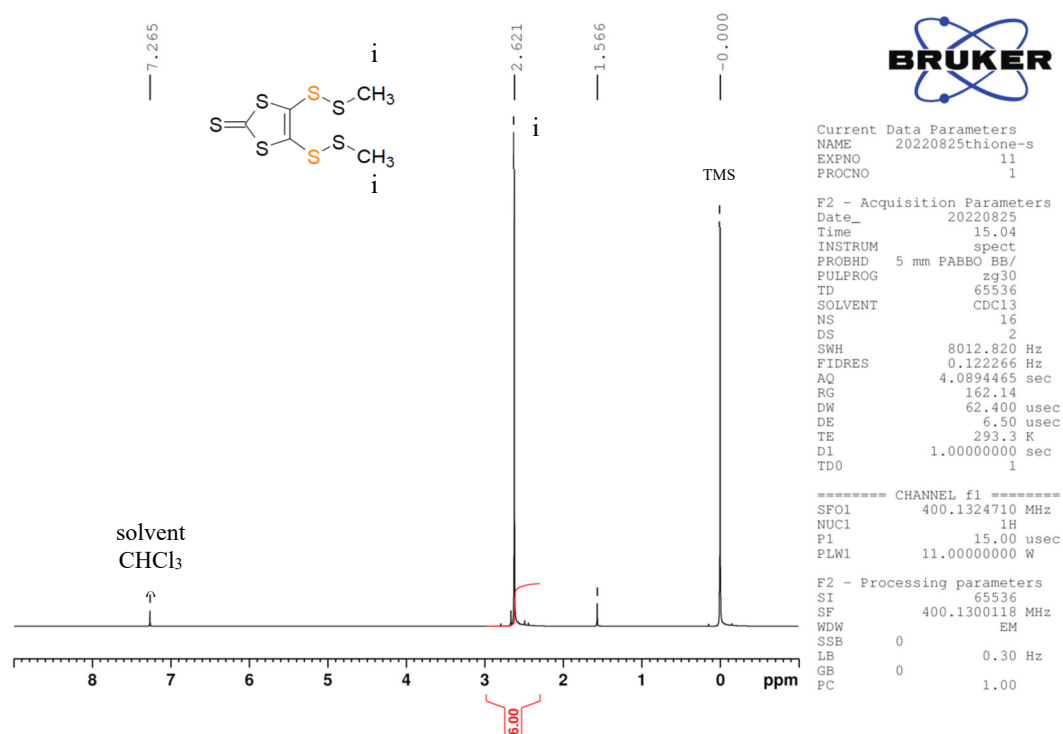
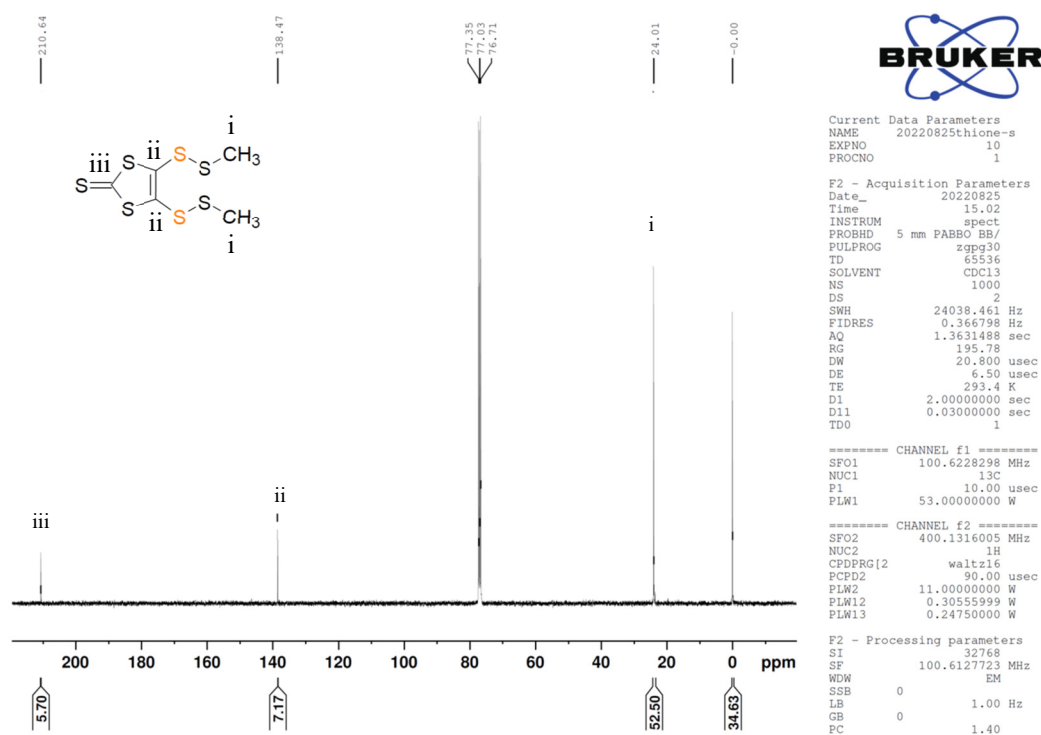
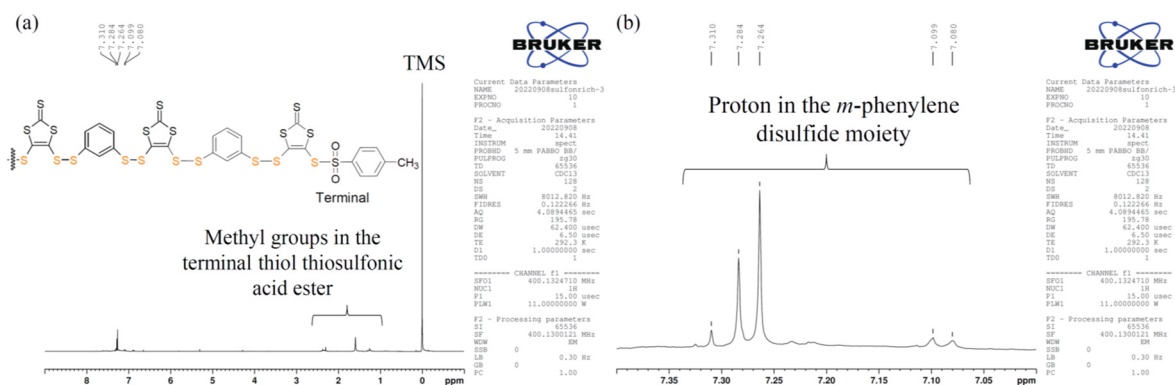
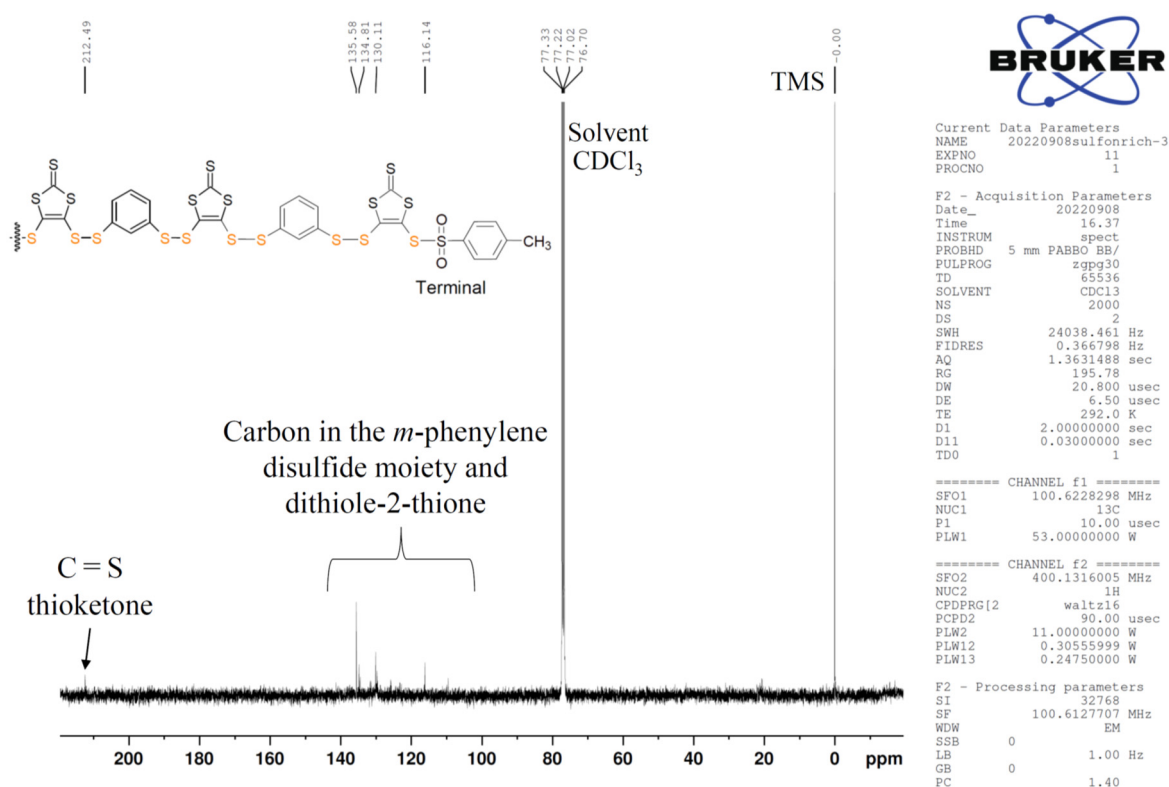


Figure S2. <sup>13</sup>C NMR chart of molecular 1.

Figure S3.  $^1\text{H}$  NMR charts of molecular **2a**, 0–8 ppm (a) and 7.15–7.85 ppm (b).Figure S4.  $^{13}\text{C}$  NMR charts of molecular **2a**, 0–160 ppm (a) and 123–140 ppm (b).Figure S5.  $^1\text{H}$  NMR chart of molecular **2b**.

Figure S6.  $^{13}\text{C}$  NMR chart of molecular 2b.

Figure S7. <sup>1</sup>H NMR chart of molecular 3'ab, 0-8 ppm (a) and 7.0-7.4 ppm (b).Figure S8. <sup>13</sup>C NMR chart of molecular 3'ab.

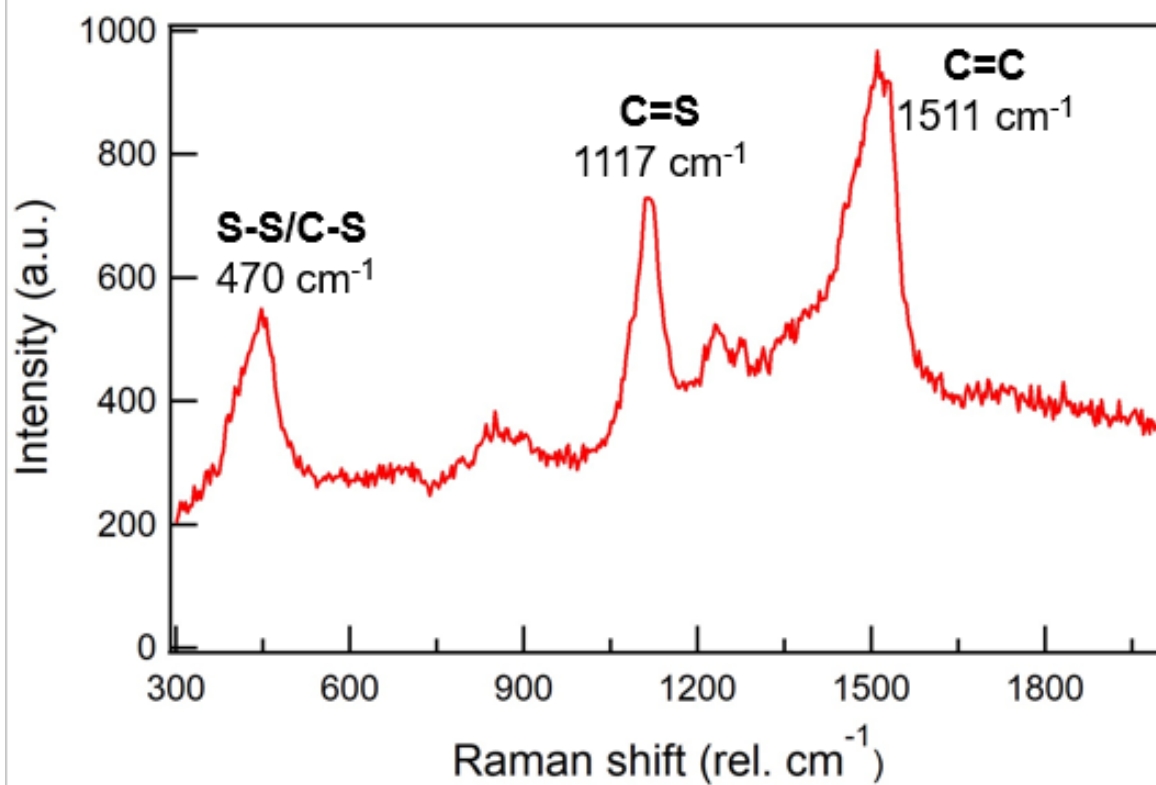


Figure S9. The Raman spectrum of 3ab.

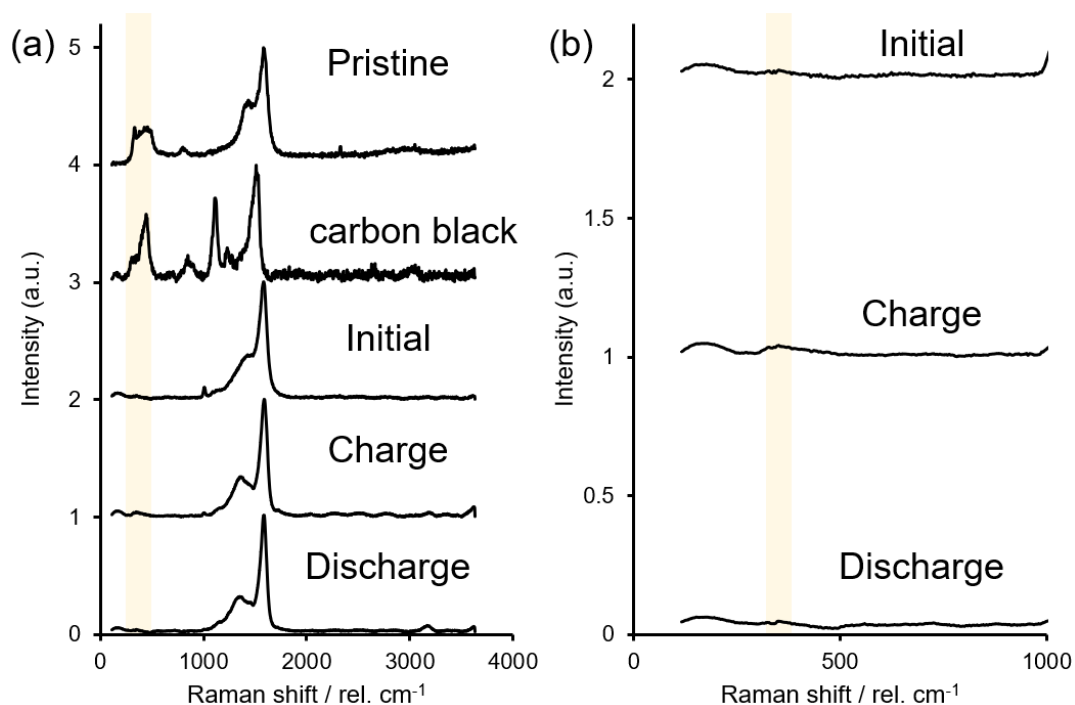
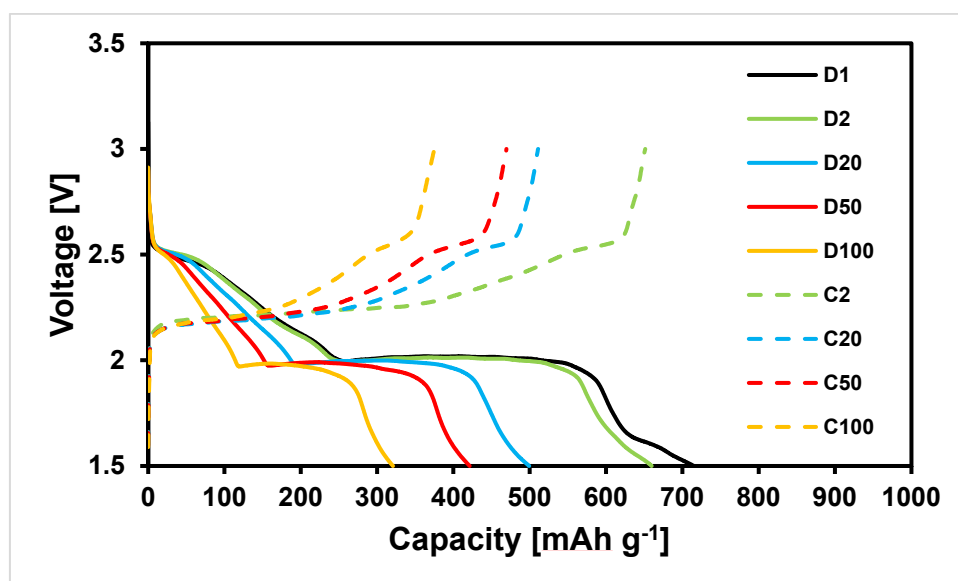


Figure S10. *Ex situ* Raman spectra of 3ab in the initial, discharge, and charge states. (a) Raman shift: 0–4000  $\text{cm}^{-1}$ , (b) 0–1000  $\text{cm}^{-1}$ .



**Figure S11.** The discharge-charge curves of the battery including 70 wt% of **3ab** (carbon agent: 20 wt%, PVDF: 10 wt%).

**Table S1.** Comparison among the capacities of the conductive polymers with sulfur chains.

Samples	Current density [mA g <sup>-1</sup> ]	Initial capacity [mAh g <sup>-1</sup> ]	Capacity retention [mAh g <sup>-1</sup> , cycles, %]	Ref
Polysulfide PPy	0.1 mA cm <sup>-2</sup>	515	452, 20, 87.8	S1
Copolymer cp (S-g-PANI)/S	167.5	1002	733, 220, 73.2	S2
Capped CP(S3BT)	1675	928	682, 500, 73.5	S3
<b>3a</b>	200	750	445, 20, 59.3	<b>This work</b>
<b>3ab</b>	200	1096	681, 20, 62.1	<b>This work</b>

## References

- S1. Shi Chao, Z.; Lan, Z.; Jinhua, Y. Preparation and Electrochemical Properties of Polysulfide Polypyrrole. *J. Power Sources* **2011**, *196*, 10263–10266.
- S2. Dai, S.; Feng, Y.; Wang, P.; Wang, H.; Liang, H.; Wang, R.; Linkov, V.; Ji, S. Highly Conductive Copolymer/Sulfur Composites with Covalently Grafted Polyaniline for Stable and Durable Lithium-Sulfur Batteries. *Electrochim. Acta* **2019**, *321*, 134678.
- S3. Zeng, S.; Li, L.; Zhao, D.; Liu, J.; Niu, W.; Wang, N.; Chen, S. Polymer-Capped Sulfur Copolymers as Lithium-Sulfur Battery Cathode: Enhanced Performance by Combined Contributions of Physical and Chemical Confinements. *J. Phys. Chem. C* **2017**, *121* (5), 2495–2503.