Supporting Information

Fast Polysulfides Catalytic Conversion and Self-Repairing Ability for High Loading Lithium-Sulfur Batteries using a Permselective Coating Layer Modified Separator

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Fig. S1 SEM images of AB powder

| Material | BET surface area | Pore diameter | Pore volume | |
|----------|------------------|---------------|---------------|--|
| | $(m^2 g^{-1})$ | (nm) | $(mL g^{-1})$ | |
| AB | 55.991 | 12.355 | 0.203 | |
| ABPS | 57.335 | 18.399 | 0.252 | |



Fig. S2 The TG curves of CPS/S composites from 50 to 600°C at a heating rate of 10 °C min⁻¹ under a N_2 atmosphere.



Fig. S3 The cross section SEM images of the sulfur cathodes with (a) 2.5 mg cm⁻² and (b) 6.8 mg cm⁻².



Fig. S4 The cross section SEM images of the AB modified separator



Fig. S5 Optical observation of Li_2S reaction with AB and ABPS composite in DOL/DME solution after 2 h.



Fig. S6 The surface morphology images of the as-prepared sulfur electrodes.

| Table S2 Parameters obtained for t_{Li} + calculation of the symmetric coin co | ell. |
|---|------|
|---|------|

| Separator | <i>I</i> _s , μΑ | $R_{ m b}, \Omega$ | R_{f}, Ω | R_0, Ω | $t_{\rm Li}^+$ |
|----------------------------|----------------------------|--------------------|--------------------------|---------------|----------------|
| Pristine Celgard separator | 92.1 | 86.1 | 96.4 | 4.5 | 0.37 |
| ABPS modified separator | 310.2 | 25.4 | 27.2 | 2.8 | 0.57 |

Table S3 Characteristics of various high loading sulfur electrodes with or without a novel functional separator reported in the literature.

| I | nterlayer or Host Material ^{Reference} | Thick of the coating layer (µm) | Mass loading of the coating (mg cm ⁻ ²) | S Percentage of Electrode/wit h coating (wt%) | S mass Loading (mg cm ⁻²) | E/S, μL mg ⁻¹ | Cycle Performance mA h g ⁻¹ (cycle numbers) | Areal Capacity, mA h cm ⁻² | Published Date |
|----------|--|---|--|---|--|-----------------------------|---|---|-------------------|
| | g-C ₃ N ₄ /CNT ^[53] | 12 | 0.45 | 57.6/55.6 | 7.2 | 6 | 787~633, (0.2C, 120) | 5.67~4.56 | 2020 |
| s | ACC/MnO2 ^[54] | 20 | - | 80/- | 5 8 | 6.25 6.25 | 993~712 (0.2C, 300) 882-686 (0.1, 100) | 4.96~3.56 7.06~4.49 | 2020 |
| | BFO/GO/AB ^[55] | 25 | 0.48 | 64/60.7 | 5.6 | 12 | 1016~ 834 (0.1C, 50) | 5.1~4.3 | 2020 |
| ayer | LNPO/rGO ^[56] | 20 | 0.88 | 80/71.6 | 6 | 8.3 | 700~623 (0.3C, 300) | 4.2~3.7 | 2020 |
| al la | FeTaPc@rGO ^[57] | 13 | 0.23 | 68/65.9 | 5 | 5 | 1000~800 (0.2C, 200) | 5~4 | 2021 |
| Function | Ce-MOF-2/CNT ^[36] | 8 | 0.4 | 80% for composite/- | 6 | 40 (for each cell) | 993.5~886.4 (0.1C, 200) | 5.96~5.32 | 2019 |
| | FeP/HPC ^[52] | | 0.3 | 52.5/51.1 | 5.73 | 15 | 1052.9~982.5 (0.01C, 30) | 6.03~5.63 | 2021 |
| | CoFeCN@C ^[27] | 15 | 0.5 | 70/65.8 | 5.5 | 15 | 982~818 (0.1C, 150) | 5.4~4.5 | 2021 |
| | Fe/Co/Ni SACs ^[58] | 7 | 0.1 | -/- | 4.5 | 10 | 1000~892 (1.0 C, 750) | 4.5~4.0 | 2019 |
| | Co-N-C/AC ^[59] | 7 | 0.5 | 56/53.1 | 5.09 | 10 | 1100 (0.2C, 100) | 5.6 | 2021 |
| | CoSA-N-C@S ^[60] | | | 55.6 | 4.9 | 10.4 | 1061.7~871.3 (0.2C, 120) | 4.63~4.21 | 2021 |
| | nO2@rGO/S ^[61] | | | 56 | 4 4 | 5 4 | 748~446 (0.2C, 200) 710~578 (0.2C, 100) | 2.99~1.78 2.84~2.31 | 2020 |
| ials | 3D-S@NCoCPC[37] | | | 62 | 6.15 | 4 | 1040~940 (100mA g ⁻¹ , 7) | 6.39~5.78 | 2021 |
| t Mater | S@3DVS ₄ @RGO ^{[62}] | | | 70 | 5 7 | 10 10 | 1116~996 (0.2C, 100) 1030~853 (0.2C, 100) | 5.80~4.98 7.21~5.97 | 2020 |
| Hos | Fe-Ni/S ^[63] | | | 73.1 | 6.4 | 8 | 953~797 (0.1C, 100) | 6.1~5.1 | 2021 |
| tional | CNT/CNF –polysulfide ^[64] | | | 68 | 8.64 8.64 | 7 4 | 880~400 (0.1C, 200) 620~0 (0.1C, 100) | 6.9~3.45 5.35~0 | 2021 |
| Fun | S@Co-N/G [65] | | | 67.5 | 6 | 12 | 1210~850 (0.2C, 100) | 7.26~5.1 | 2019 |
| | G-MgB ₂ ^[28] | | | 60 | 9.3 | 6.5 | 850~665 (0.2C, 100) | 7.9~6.18 | 2019 |
| | FLPT-S ^[66] | | | 59 | 6.8 | 8 | 1120~805 (1/30C, 75 cycles) | 7.62~5.48 | 2019 |

| | ABPS-coating layer ^{This work} | 5.5 | 0.12 | 72/69.6 | 6.8 | 4 | 1211~886 (1.03 mA cm ⁻² , 170) | 8.24~6.03 | This work |
|--|--|-----|------|---------|-----|---|---|-----------|-----------|
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