

### *Supporting Information*

## **A multifunctional polyimide nanofiber separator with a self-closing polyamide-polyvinyl alcohol top layer of Turing structure for high-performance lithium-sulfur batteries**

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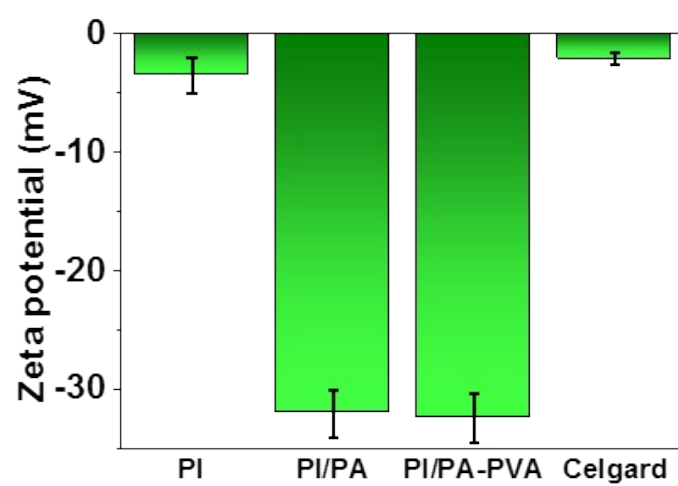
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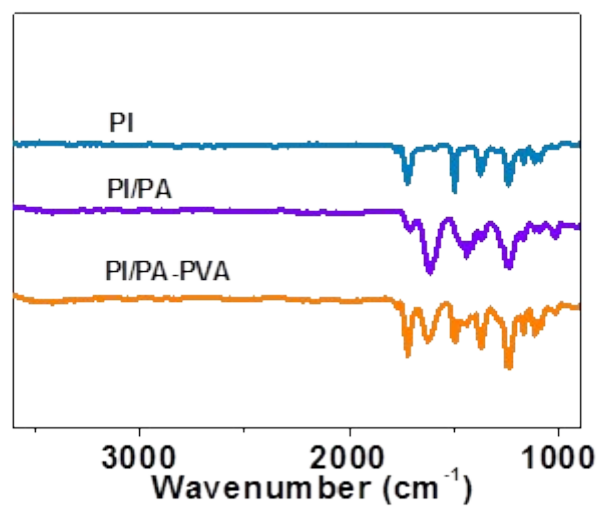
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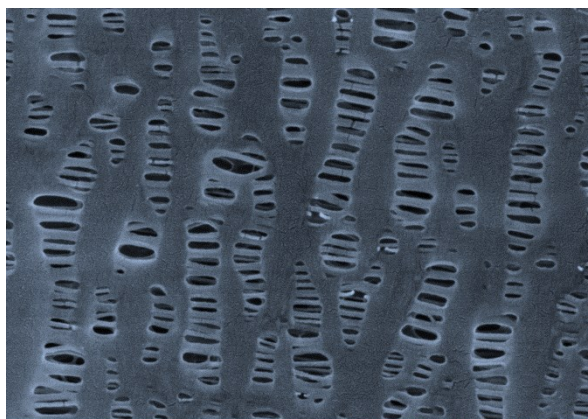
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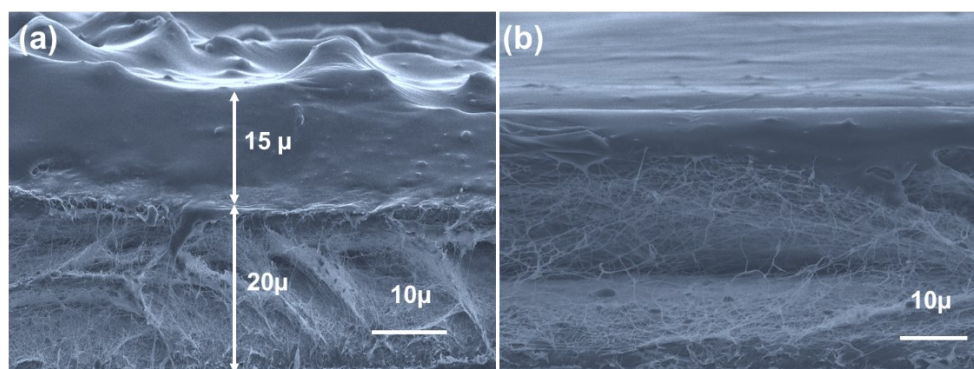
**Figure S1.** Surface zeta potentials of different separators.



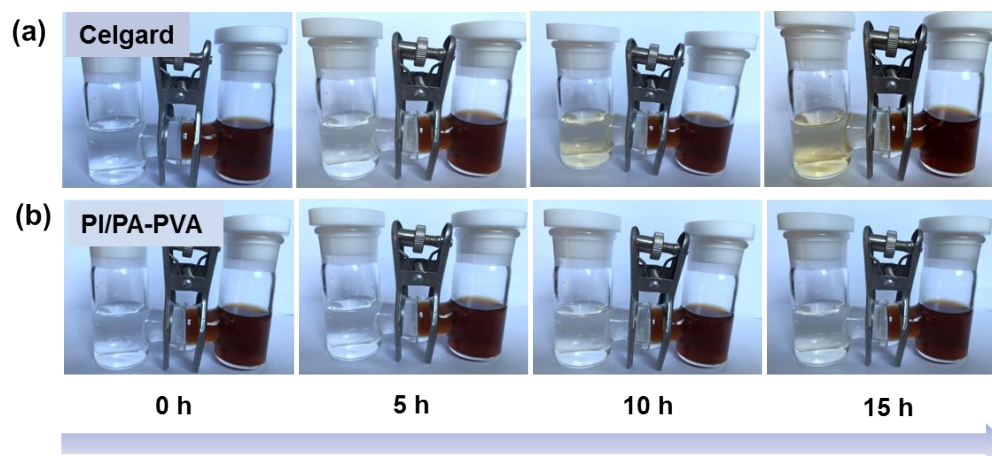
**Figure S2.** FTIR spectra of different separators.



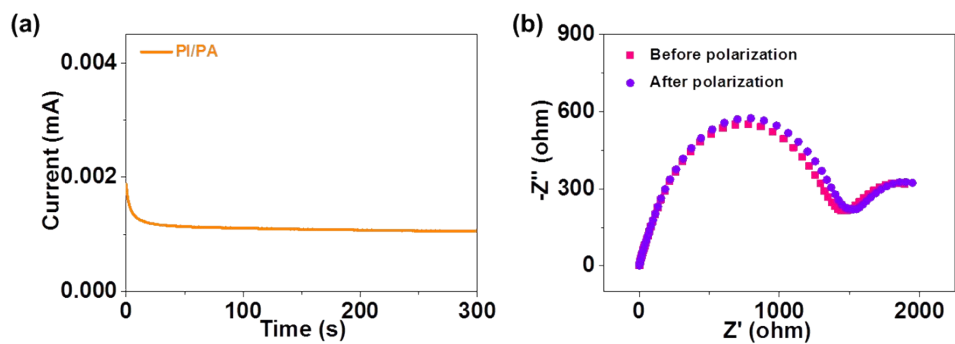
**Figure S3.** SEM image of the Celgard separator.



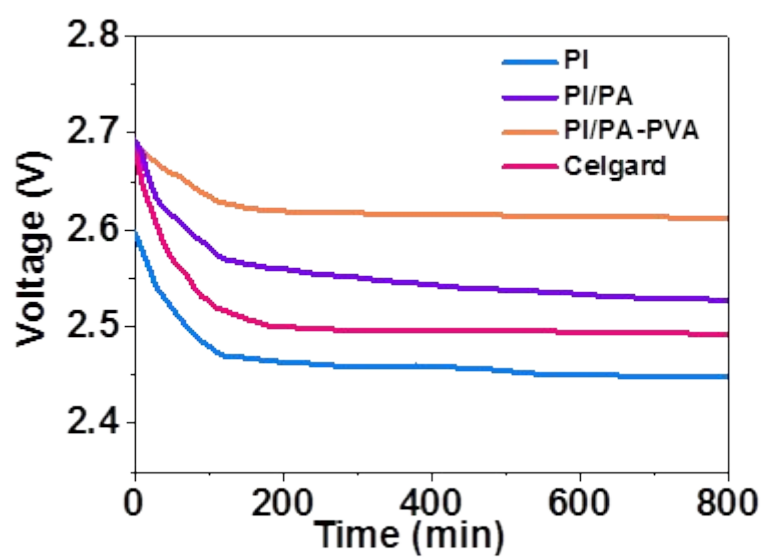
**Figure S4.** SEM images of cross-section of PI/PA-PVA separator: (a) fresh prepared, (b) after heating at 230 °C.



**Figure S5.** Photographs showing the polysulfide diffusion across the Celgard (a) and PI/PA-PVA (b) separators for different time, respectively.

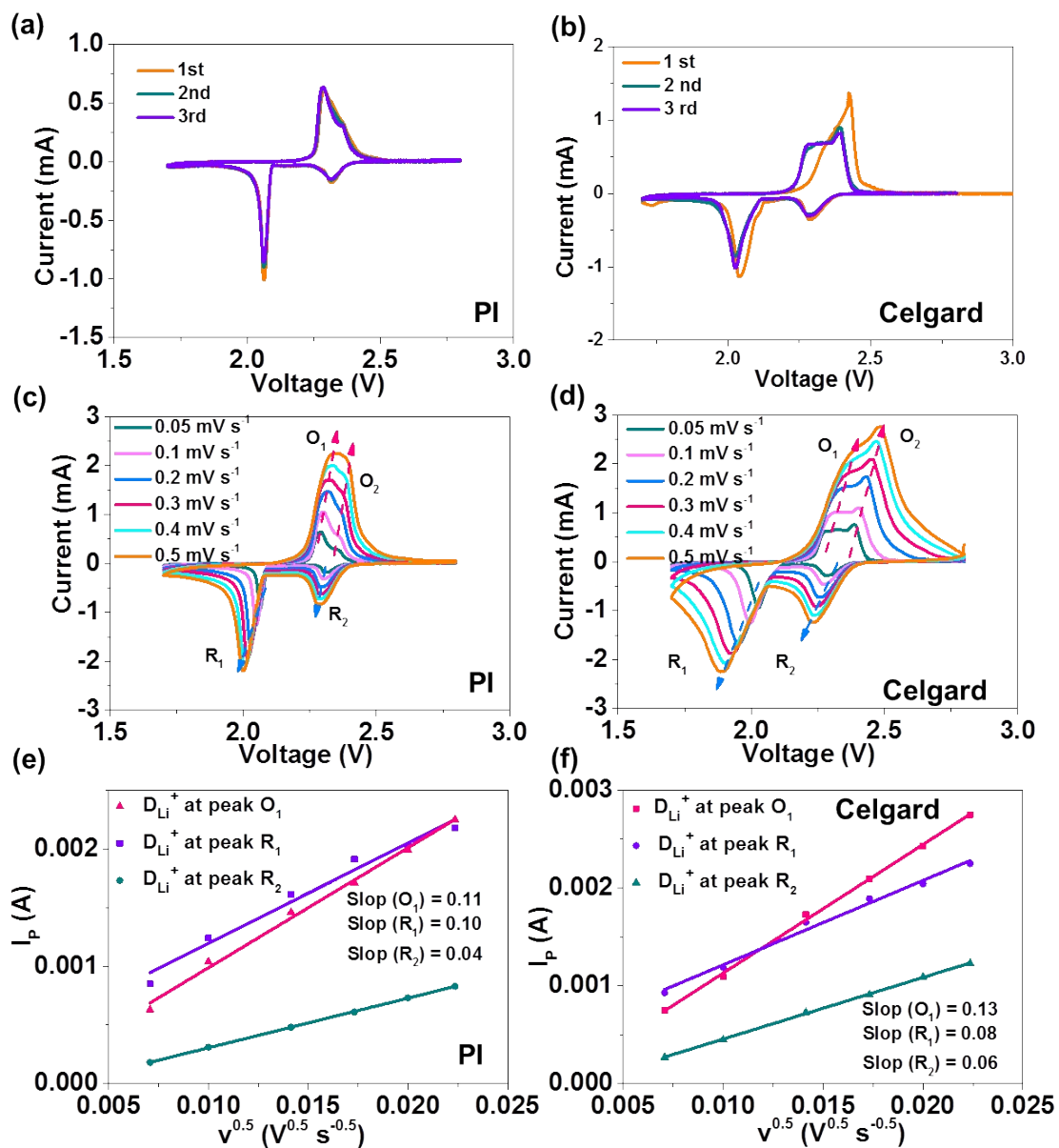


**Figure S6.** (a) Chronoamperometry profile of the PI/PA separator and (b) the corresponding impedances under the initial and steady-state current conditions.

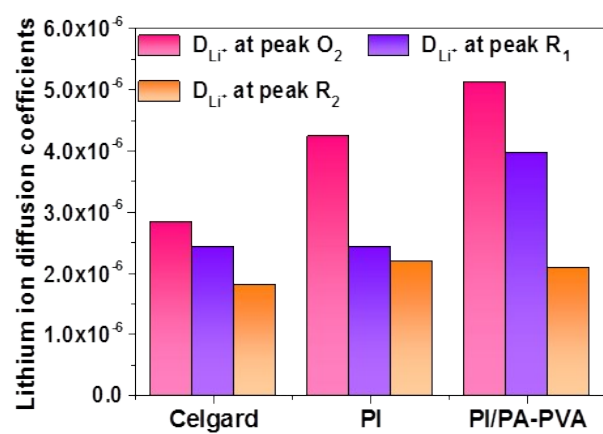


**Figure S7.** Open circuit voltage profiles showing the self-discharge behavior of Li-S batteries with PI, PI/PA-PVA and Celgard separators.

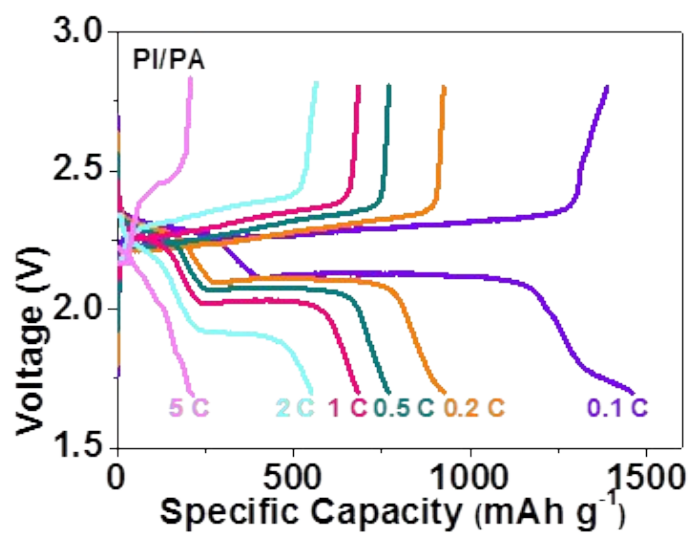




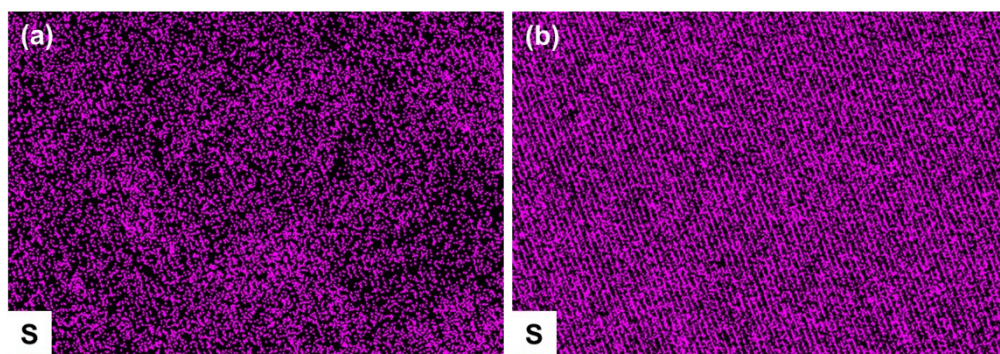
**Figure S8.** (a-b) CV curves of Li-S batteries with PI and Celgard separators at a scanning rate of 0.1 mV s<sup>-1</sup>, (b-d) CV curves of Li-S batteries with PI and Celgard separators under different scanning rates and (e-f) the corresponding linear fits of the peak currents.



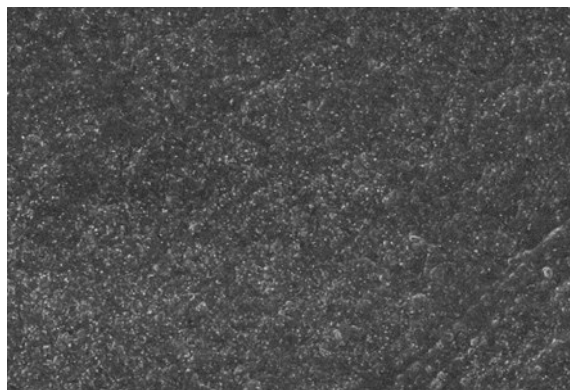
**Figure S9.** Lithium ion diffusion coefficients for different separators



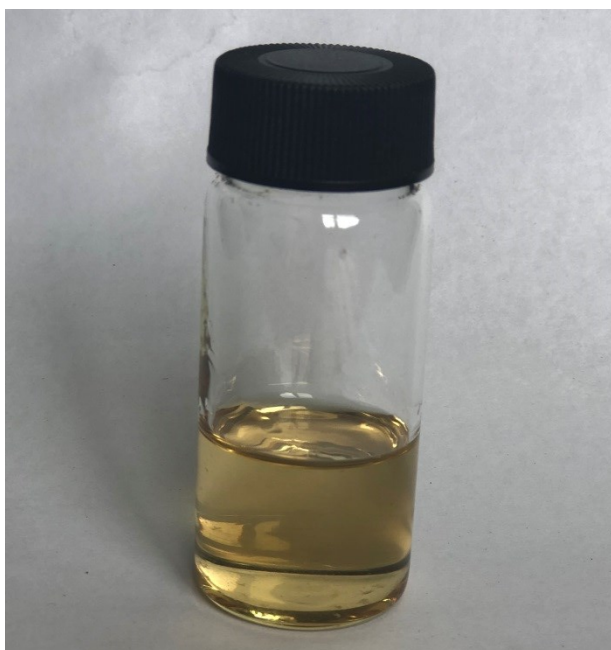
**Figure S10.** Discharge-charge profiles of Li-S batteries with the PI/PA separator at different rates.



**Figure S11.** Elemental mappings of sulfur on the cathode faced with (a) Celgard and (b) PI/PA-PVA separators after rate test.



**Figure S12.** SEM image of the pristine lithium anode.



**Figure S13.** Photograph of the synthesized PAA in NMP solution (15%).

**Table S1.** Physical properties of different separators.

| Membrane  | Thickness ( $\mu\text{m}$ ) | Porosity (%)   | Contact angle ( $^\circ$ ) | Electrolyte uptake (%) | Electrolyte retention (%) |
|-----------|-----------------------------|----------------|----------------------------|------------------------|---------------------------|
| PI        | $30 \pm 0.5$                | $92.1 \pm 0.5$ | 0                          | 2011                   | 70.2                      |
| PI/PA     | $30 \pm 0.5$                | $76.6 \pm 0.5$ | 35.8                       | 1321                   | 41.1                      |
| PI/PA-PVA | $30 \pm 0.5$                | $75.1 \pm 0.5$ | 0                          | 1640                   | 52.6                      |
| Celgard   | $25 \pm 0.5$                | $41.3 \pm 0.5$ | 49.2                       | 156                    | 11.5                      |

**Table S2.** Electrochemical performance comparison of this work with the previous reports involving different separators using carbon-sulfur cathodes in Li-S batteries.

| Separator                     | Sulfur    | Rate                         |                             | Fading rate per cycle (%)      | Refs             |
|-------------------------------|-----------|------------------------------|-----------------------------|--------------------------------|------------------|
|                               | (%)       | Initial capacity             | capability                  |                                |                  |
|                               |           | [mA h g <sup>-1</sup> ]      | [mA h g <sup>-1</sup> ]     |                                |                  |
|                               |           | (Current rate)               | (Cycling rate)              |                                |                  |
| CNT/PP <sup>a</sup>           | 60        | 1179(0.1 C)                  | 430 (2 C)                   | 0.4 (100 cycles, 0.1C)         | 1                |
| Black phosphorus/Celgard      | 80        | 930 (0.4 A g <sup>-1</sup> ) | 623(3.5 A g <sup>-1</sup> ) | 0.14 (100 cycles, 0.1 C)       | 2                |
| AC/GF <sup>b</sup>            | 70        | 1324 (0.2 C)                 | 362 (1.5 C)                 | 0.8 (50 cycles, 0.2 C)         | 3                |
| PAA-SWNT/Celgard <sup>c</sup> | 65        | 1130(0.1 C)                  | 592 (2 C)                   | 0.13 (100 cycles, 0.2 C)       | 4                |
| TiN/Celgard                   | 60        | 1061 (0.5 C)                 | 329 (2 C)                   | 0.15 (250 cycles, 0.5 C)       | 5                |
| MEC-AA <sup>d</sup>           | 70        | 915 (0.5 C)                  | 720 (1 C)                   | 0.2 (150 cycles, 0.5 C)        | 6                |
| KB@Ir/Celgard <sup>e</sup>    | 75        | 1600 (0.1 C)                 | 653 (2 C)                   | 0.25 (100 cycles, 0.2C)        | 7                |
| MoS <sub>2</sub> /Celgard     | 65        | 1471 (0.1 C)                 | 550(1 C)                    | 0.08 (600 cycles, 0.5C)        | 8                |
| SiO <sub>2</sub> /PP          | 60        | 937 (0.2 C)                  | 621 (1 C)                   | 0.23 (200 cycles, 0.2 C)       | 9                |
| <b>PI/PA-PVA</b>              | <b>70</b> | <b>1499 (0.1 C)</b>          | <b>497 (5 C)</b>            | <b>0.1 (500 cycles, 0.2 C)</b> | <b>This work</b> |

<sup>a</sup> PP: Polypropylene

<sup>b</sup> AC/GF: Biomass-derived porous carbon / glass fiber

<sup>c</sup> PAA-SWNT/Celgard: Poly(acrylic acid) coated single-walled carbon nanotube film on Celgard.

<sup>d</sup> MEC-AA: MCM-41/ carbon nanotubes wrapped poly-(ether imide) nanofibers

<sup>e</sup> KB@Ir/Celgard: Ketchen Black and Ir nanoparticle modified Celgard



**Table S3.** Specific conditions of the interfacial polymerization for different samples.

| Samples   | Monomer concentration |                              |                             |
|-----------|-----------------------|------------------------------|-----------------------------|
|           | TMC (mM)              | PZ (mM)                      | PVA (mM)                    |
| PI        | 60                    | 1179(0.1 C)                  | 430 (2 C)                   |
| PI/PA     | 80                    | 930 (0.4 A g <sup>-1</sup> ) | 623(3.5 A g <sup>-1</sup> ) |
| PI/PA-PVA | 70                    | 1499 (0.1 C)                 | 497 (5 C)                   |

## References

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