# Supporting Information

# Polysulfides Mitigation through Tailored Separator for Critical Temperature Energy-Dense Lithium-Sulfur Batteries

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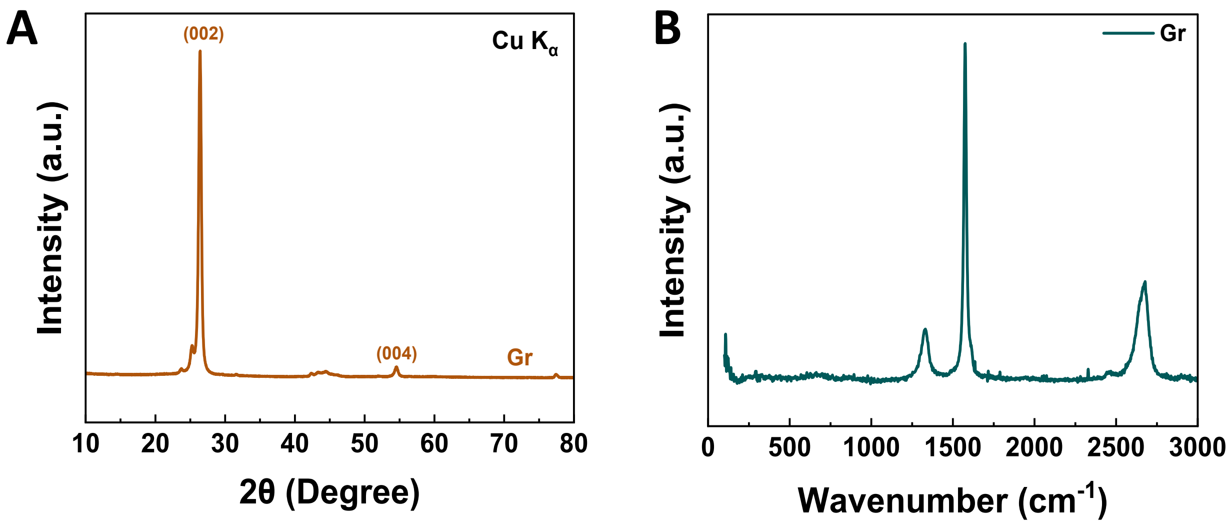


**Figure S1:** A: XRD pattern and B: Raman Spectrum of Graphene.

The structures and properties of the graphene used for the tailored separator were characterized via X-ray diffraction and Raman. The XRD result (Figure S1-A) shows that there are two characteristic peaks in the XRD pattern of the graphene (around $2\theta = 26.44°$ and $2\theta = 54.56°$), with an interlayer spacing of <4 Å (calculated using Bragg's law, $\lambda = 2d\sin(\Theta)$) can accommodate polysulfides to suppress shuttling effect across the separator. As shown in Figure S1-B, Raman peaks at 1279 $cm^{-1}$, 1530 $cm^{-1}$, and 2677 $cm^{-1}$ are graphene's characteristic peaks, which can be attributed to the D, G, and D′(2D) bands, respectively.

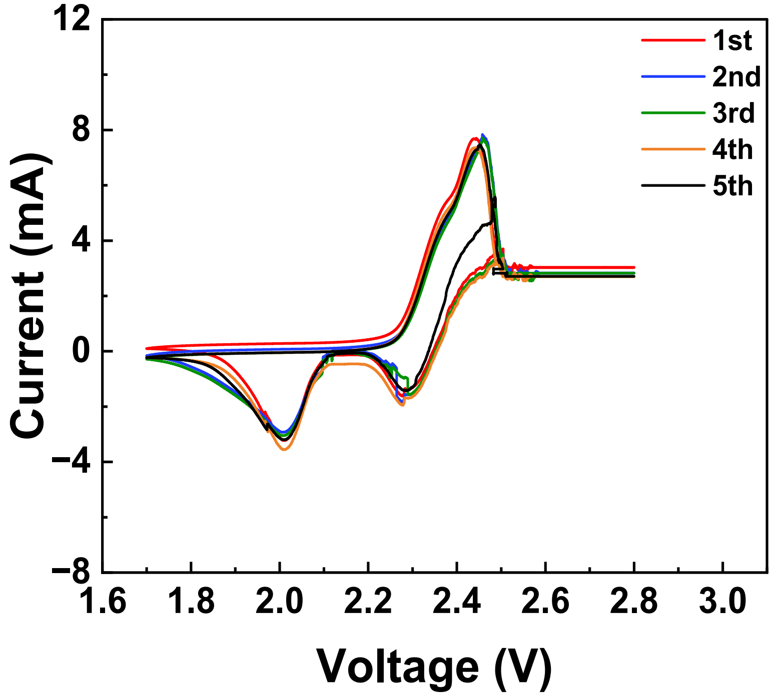


**Figure S2:** Cyclic voltammetry of Li-S cell with pristine separator at 25 °C.

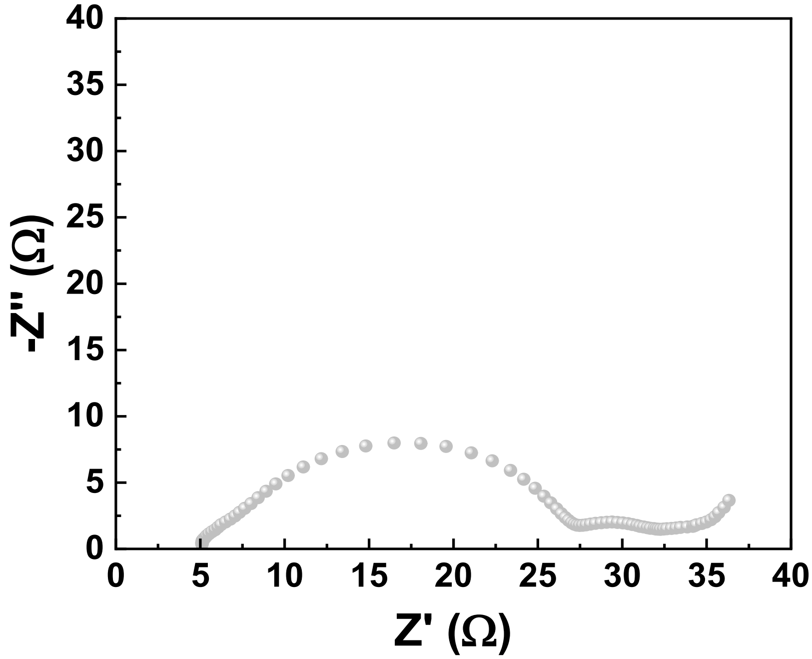


**Figure S3:** EIS of Li-S cell with conventional PP separator.

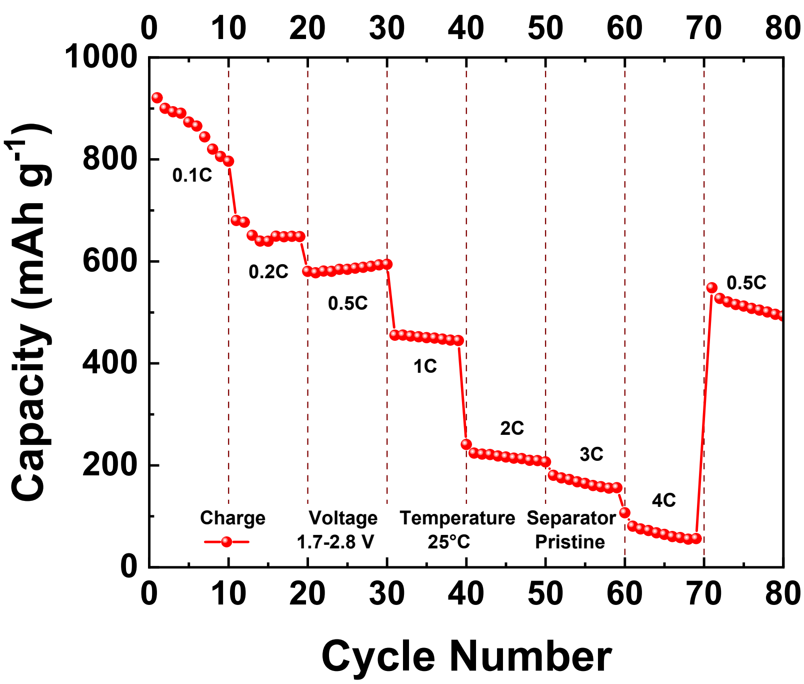


**Figure S4:** Rate studies exhibiting performance at 0.1C, 0.2C, 0.5C, 1C, 2C, 3C, and 4C of Li-S cell with pristine separator at 25 °C.

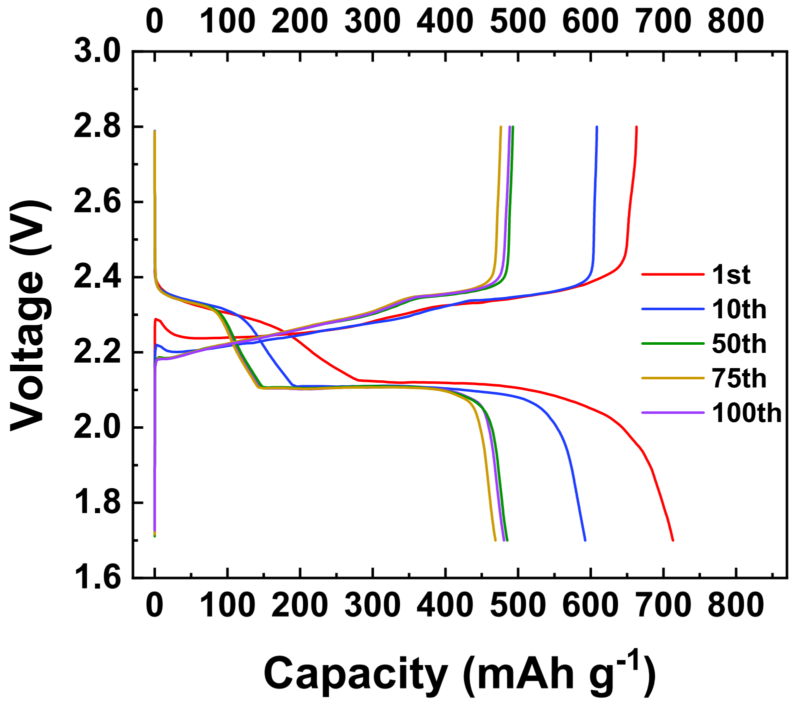


**Figure S5:** Voltage characteristics of Li-S cell with pristine separator at 25 °C.

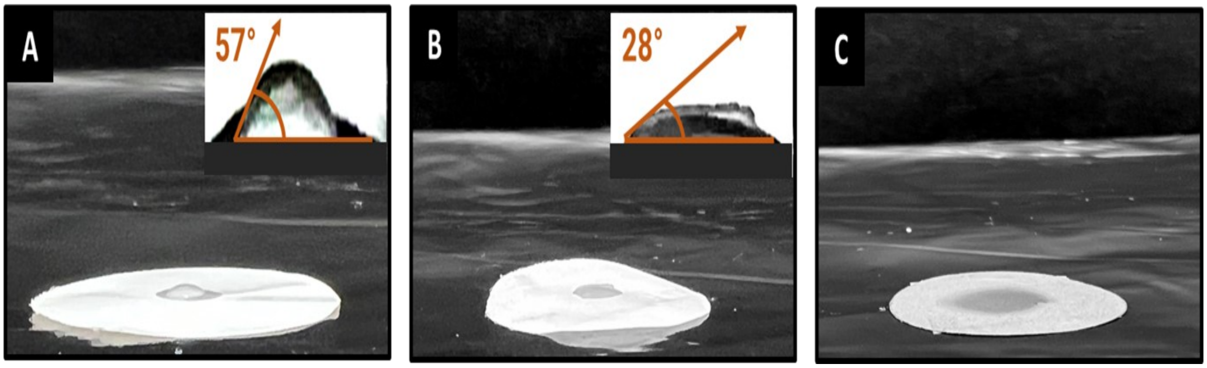


**Figure S6**: Photographs showing the wetting behavior of separators with electrolyte. A: Pristine PP separator; B: PDA coated separator with insets of surface contact angle measurement; C: Completely wetted tailored separator consisting of Graphene PDA coating.

The wettability tests on separators were performed via the surface contact angle measurement method with electrolyte as shown in Figure S6. The PDA coating reduces the surface tension and improves the wettability of the electrolyte, reflecting in a smaller contact angle (28°) in comparison with pristine PP separator (57°). Whereas superior electrolyte wettability is achieved by graphene-coated tailored separator with higher porosity improving $Li^+$ transportation in the electrolyte, and thus enhancing the electrochemical performance of the battery.

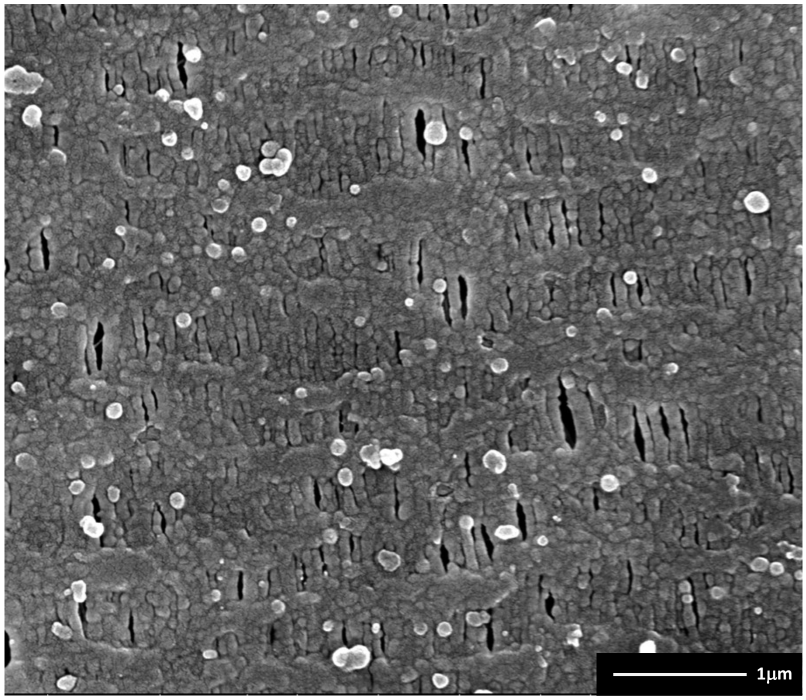


**Figure S7:** SEM Top view of PDA coated PP separator

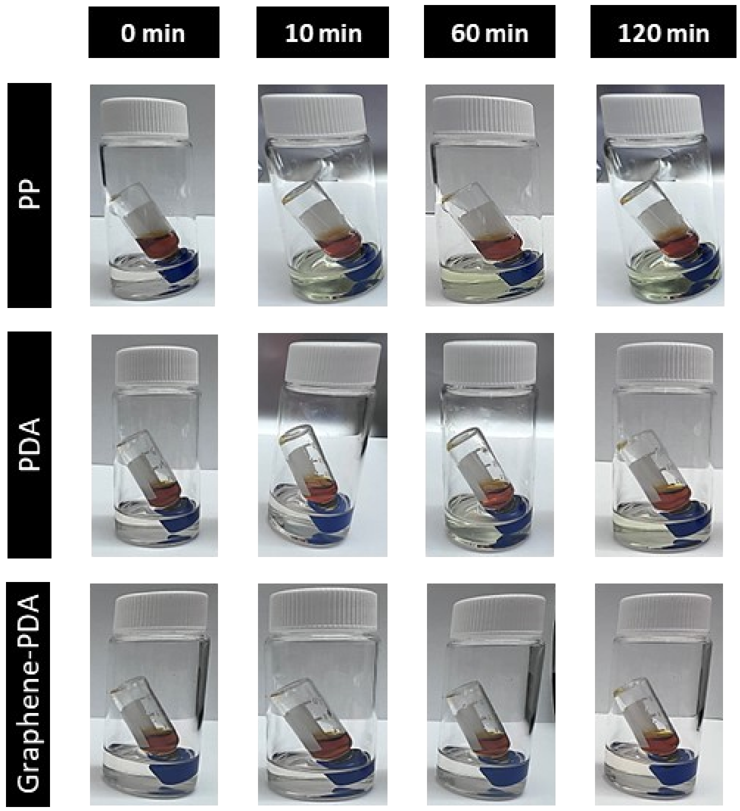


**Figure S8:** Visual demonstration of polysulfide diffusion tests of the A: Pristine PP separator; B: PDA coated separator; and C: tailored separator consisting of Graphene PDA coating after various resting times (10, 60, and 120 minutes) using diffusion test bottles.

Visual verification of the polysulfide diffusion test is demonstrated in Figure S8 using diffusion test bottles, where polysulfide solution (0.2 M $Li_2S_6$/DME+DOL) and blank solvent (DME/DOL) were separated by separator for various resting times. The dissolved polysulfide could rapidly diffuse through the Pristine PP separator, since the colorless DME/DOL solution turned pale yellow after only 10 minutes of rest, and the color intensified over time, implying that the pristine separator is ineffective in curbing the polysulfide diffusion across a separator. In contrast, PDA coated separator could significantly reduce the intensity of diffusion but is not enough to completely mitigate polysulfide. On the other hand, the Graphene-PDA coated tailored separator enables a clear and colorless solution even after 120 mins, confirming its great capability of blocking polysulfide diffusion and stabilizing the Li-S battery chemistry.