Supporting Information

Rational Design of a Gel-polymer-inorganic Separator with Uniform Lithium-ion Deposition for Highly Stable Lithium-Sulfur Batteries

Pengfei Wang a, Jiejun Bao a, Kezhong Lv a, Na Zhang a, Zhi Chang b, Ping He a*,
Haoshen Zhou a,b*

a Center of Energy Storage Materials & Technology, College of Engineering and Applied Sciences, Jiangsu Key Laboratory of Artificial Functional Materials, National Laboratory of Solid State Microstructures, and Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210093, PR China

b Energy Technology Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), 1-1-1, Umezono, Tsukuba 305-8568

* Corresponding author: Ping He
E-mail: pinghe@nju.edu.cn

* Corresponding authors: Haoshen Zhou
E-mail: hszhou@nju.edu.cn
Polysulfides absorbent:

**Fig. S1.** Schematic illustration of interaction mechanism between polysulfides and CMC.
Fig. S2. The cross-section of PP separator (a), Al₂O₃-PE separator (b) and Al₂O₃-PE-CMC separators with different thickness of CMC coating layer (c-f).
Fig. S3. SEM images of (a) Pure S, (b) Ketjenblack, (c, d) S@Ketjenblack.
Fig. S4. The TG curve of S@Ketjenblack (a), XRD patterns of S, Ketjenblack and S@Ketjenblack (b), TG-DSC curves of PP (c) and Al₂O₃-PE (d) separator in air atmosphere.
Fig. S5. XRD patterns of different side of PP, PE-Al$_2$O$_3$ and CMC-PE-Al$_2$O$_3$ separators.
Fig. S6. SEM images of CMC film on the Al₂O₃-PE-CMC separator at different magnifications with the corresponding scale bar of 2 μm (a) and 500 nm (b).
Fig. S7. The thermal stability tests for polymer and Al₂O₃-coated polymer separator (a) pristine, (b) after 120 °C for 1h and (c) after 150 °C for 1h.
Fig. S8. Cycling performances and coulombic efficiency with high S loading of the PP, Al₂O₃-PE and Al₂O₃-PE-CMC separators in Li-S full cells (a) and Charge/discharge curves of Al₂O₃-PE-CMC separator (b).
**Fig. S9.** Polysulfide permeation measurements for the PP (a), Al₂O₃-PE (b) and Al₂O₃-PE-CMC (c) separators during the course of polysulfide diffusion from the right side to the left side of the H-shaped glass bottles, the Raman spectra of the pure TEGDME and the polysulfide solution in the left side after 12 h (d).
**Fig. S10.** Polysulfide permeation measurement for the PP separator after (a) 2 days, (b) 3 days, (c) 4 days and (d) 5 days.
Fig. S11. Specific fitting results from the EIS of PP (a), Al_2O_3-PE (b) and Al_2O_3-PE-CMC (c) separators in Li-S full cells.
Fig. S12. The structure schematic of the battery using Al₂O₃-PE-CMC separator anchored with polysulfides and C cloth as cathode (a), the corresponding cycling performance (b), charge/discharge curves (c) and differential capacity curves (d).