Supporting Information

Mosaic rGO Layers on Lithium Metal Anodes for Effective Mediation of Lithium Plating and Stripping

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Fig. S1. Sequential photos for illustrating the process of preparing rGO-coated Li disks. (a) An aqueous solution of graphite oxide. (b) A homogeneous rGO dispersion. (c) Self-assembled rGO membrane driven by thermal evaporation. (d) rGO membranes suspended in ethanol. (e) rGO membrane stored in DEC. (f) A rGO-coated lithium disk.



Fig. S2. AFM images of rGO membranes with different thickness.



0.02 mg ml⁻¹ 0.05 mg ml⁻¹ 0.1 mg ml⁻¹ 0.15 mg ml⁻¹ 0.2 mg ml⁻¹

Fig. S3. Photos of rGO membranes with different thickness on quartz plates. These rGO films were prepared by varying the concentrations of rGO solutions at (a) 0.02 mg ml⁻¹, (b) 0.05 mg ml⁻¹, (c) 0.1 mg ml⁻¹, (d) 0.15 mg ml⁻¹, and (e) 0.2 mg ml⁻¹ for 1-hour heating under 90 °C.



Fig. S4. AFM characterization of the surface roughness for rGO membranes of (a) 16 nm, (b) 65 nm, and (c) 180 nm in thickness.



Fig. S5. Characterizations of the rGO membrane (16nm). (a) Top-view AFM image. (b) Raman and (c) FTIR spectra of the rGO film.



Fig. S6. SEM images of the rGO membrane on glass taken out from different solvents showing a much flattened morphology after the evaporation of organic solvents. (a) the rGO membrane directly taken out from the water/air interface. (b) the rGO membrane taken out from DEC.



Fig. S7. Optical images of gLi and Li exposed to air.



Fig. S8. A digital image showing the size of rGO sheets prepared by the SEASA method is only limited by the container dimension.

Streamlined Operation for gLi Fabrication



Fig. S9. The streamlined operation for the fabrication of 2D rGO-coated lithium foils on a wet bench for potential scale-up.



Fig. S10. Galvanostatic cycling of the asymmetric gLi||Li battery. (a) The cycling performance of the gLi||Li cell vs. Li||Li cell at the current density of 1 mA cm⁻² for a cycling capacity of 1 mAh cm⁻². (b) Magnified voltage platforms of the gLi||Li cell at different cycling time.



Fig. S11. Top-view SEM images of both electrodes of the asymmetric gLi||Li cell after 500 cycles (1 mA cm⁻², 1 mAh cm⁻²). (a) The gLi electrode. (b) The bare Li electrode.



Fig. S12. (a) Galvanostatic cycling of the symmetric gLi||gLi cell under the current density of 10 mA cm⁻² for a cycling capacity of 2 mAh cm⁻². (b, c) SEM images of the anode and cathode surfaces after 200 cycles.



Fig. S13. EIS tests of the symmetric gLi||gLi and Li||Li cells. (a) EIS spectra of the gLi||gLi cell taken at different cycles. (b) EIS spectra of the Li||Li cell taken at different cycles.



Fig. S14. Galvanostatic cycling of the symmetric gLi||gLi cell with a thicker rGO coating of 180 nm. (a) The cycling performance of the gLi||gLi vs. Li||Li cells at the current density of 1 mA cm⁻² for a cycling capacity of 1 mAh cm⁻². (b) Magnified voltage platforms of the gLi||gLi cell at different cycling time.



Fig. S15. Top-view SEM images of both electrodes in the gLi||gLi cell with a thicker rGO coating of 180 nm after 500 cycles $(1 \text{ mA cm}^{-2}, 1 \text{ mAh cm}^{-2})$ showing less-cracked rGO. (a) The cathode side. (b) The anode side.

Fig. S16. Li plating and stripping tests with 10 galvanostatic cycles in between to quantify the Li expense and CE of gLi||Cu and Li||Cu half cells.

Fig. S17. SEM-EDX characterization of the gLi electrode after 500 cycles (1 mA cm⁻², 1 mAh cm⁻²).

Fig. S18. gLi electrodes subjected to the overcharge tests of different periods. (a) The gLi cathode disassembled from the symmetric cell after 100 cycles (1 mA cm⁻², 1 mAh cm⁻²). (b) The gLi cathode disassembled from the symmetric cell further discharged for 1 h after 100 cycles. (c) The gLi cathode disassembled from the symmetric cell further discharged for 5 h after 100 cycles. (d) The gLi cathode disassembled from the symmetric cell further symmetric cell further discharged for 10 h after 100 cycles.

Fig. S19. Top-view SEM images of (a) the gLi electrode in a SPAN-KB||gLi cell and Li electrode in a SPAN-KB||Li cell after cycled at 0.2C for 400 cycles.

Fig. S20. Electrochemical performance of Li-S batteries fabricated with the sandwich Cu/gLi anode with only 150% oversized lithium. (a, b) Cycle performance and charge/discharge profiles of the SPAN-KB||Cu/gLi coin cells at 1 C-rate (1.0 - 3.0 V). (c, d) Cycle performance and charge/discharge profiles of the SPAN-KB||Cu/Li coin cells at 1 C-rate (1.0 - 3.0 V). (e) Photos of the Cu/gLi and Cu/Li anodes disassembled from the coin cells after 50 galvanostatic cycles at 1 C.

Table 1. Comparison of symmetric cell performance for the rGO passivation coating in

 this work *vs.* other Li anode protection methods

Sample	Current density (mA cm ⁻²)	Cycling performance (overpotential @ cycles)
gLi (this work)	1	8.3 mV for 600 cycles
	5	87.9 mV for 600 cycles
	10	160 mV for 200 cycles
Layered Li-rGO ¹	1	40mV for 100 cycles
	3	100mV for 100 cycles
TG-Li ²	0.5	17mV for 1000 h
FEC-Li ³	5	200 mV for 25 h
SR-G-Li ⁴	1	9 mV for 1000 cycles
	3	32 mV for 1000 cycles
	5	47 mV for 1000 cycles
MoS2-coated Li ⁵	1	10 mV for 100 h
	3	15 mV for 100 h
EP-LiF ⁶	1	30 mV for 600 cycles
	3	90 mV for 600 cycles
	5	150 mV for 600 cycles
LiPAA-Li ⁷	0.5	50 mV for 700 h
	1	100 mV for 250 h
LIA ⁸	1	50 mV for 900 h
	3	100 mV for 300 h
	8	400 mV for 60 h
3D composite Li ⁹	1	50 mV for 100 cycles
	3	150 mV for 100 cycles
polished Li anodes ¹⁰	3	25 mV for 800 cycles
	5	50 mV for 400 cycles

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Supplementary Video 1. Manipulation of the rGO membrane in DEC.

Supplementary Video 2. Coating of the rGO membrane on a Cu foil.

Supplementary Video 3. Coating of the rGO membrane on a Li foil.