Supporting Information

The Microwave Plasma Torch Chemical Vapor Deposition of Graphene Layers on Copper Foils for Facilitating Uniform Lithium Deposition in Lithium Metal Batteries

Hao-Yu Ku[1], Yun Ku[1], Chi-Yu Lai[1], Yi-Ting Lu[1,3], Hsiang Sheng Wei[1], Hung-Hsin Shih[2], Kun-Ping Huang[2], Chi-Chang Hu[1]*

[1]Department of Chemical Engineering, National Tsing Hua University, 101, Section 2, Kuang-Fu Road, Hsin-Chu 300044 Taiwan.

[2]Division of Green Manufacture Technology, Mechanical and Systems Research Laboratories, Industrial Technology Research Institute, 195, Sec. 4, Chung Hsing Rd., Chutung, Hsinchu, 310401 Taiwan.

[3]Department of Chemical Engineering, Ming Chi University of Technology, New Taipei City, 243303, Taiwan.

*Corresponding Author: Chi-Chang Hu, NTHU Chair Professor
Department of Chemical Engineering
National Tsing Hua University
Hsin-Chu 300044, Taiwan

This supporting information includes **9 Figures** and **1 Table**.

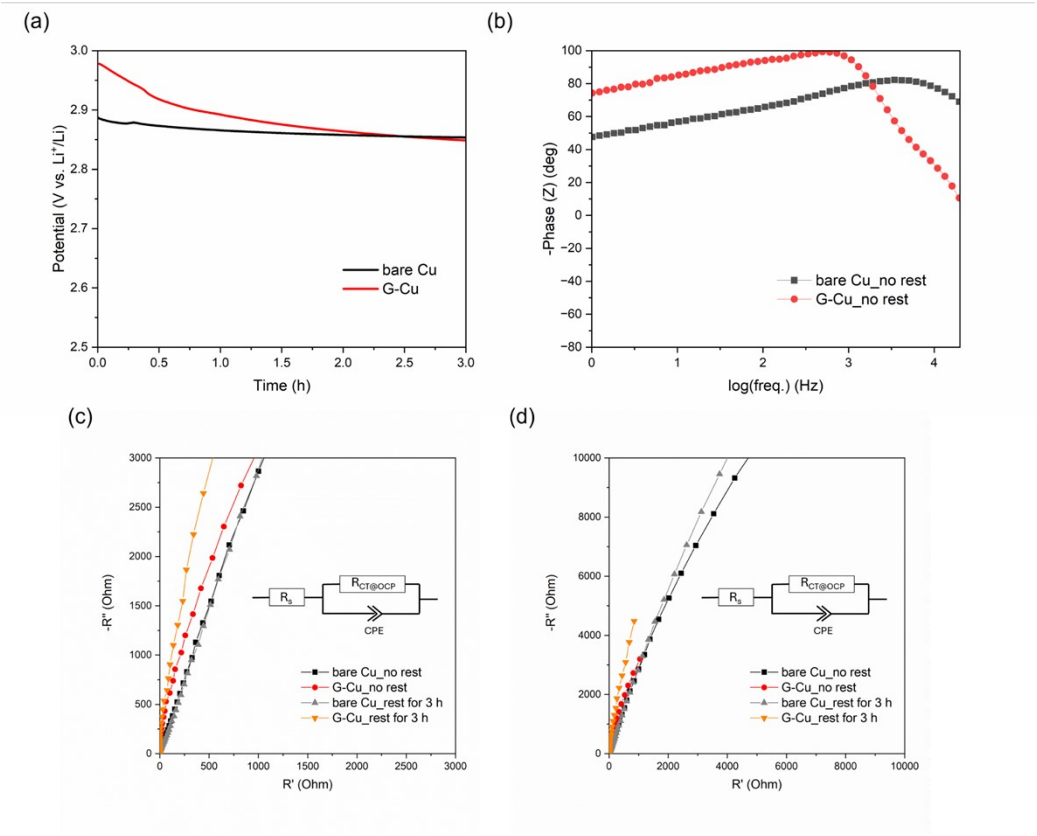

Fig. S1. (a) The variation of open circuit potentials (E_{OCP}) for the bare Cu and G-Cu foils. (b) The negative phase angle Bode plot. (c) The enlarged and (d) full Nyquist plots of the cells using bare Cu and G-Cu foils measured at their respective OCPs in the three-electrode Cu//Li cells with or without a 3-h rest.

Table S1. The fitting values of all elements in the equivalent circuit for the negative

Nyquist plots shown in Fig. S1c-d for the cells without a rest.

	Bare Cu	G-Cu
R_S (Ω)	0.69	0.48
$R_{CT@OCP}$ (Ω)	84704	20032
CPE (μF)	21.24	74.61

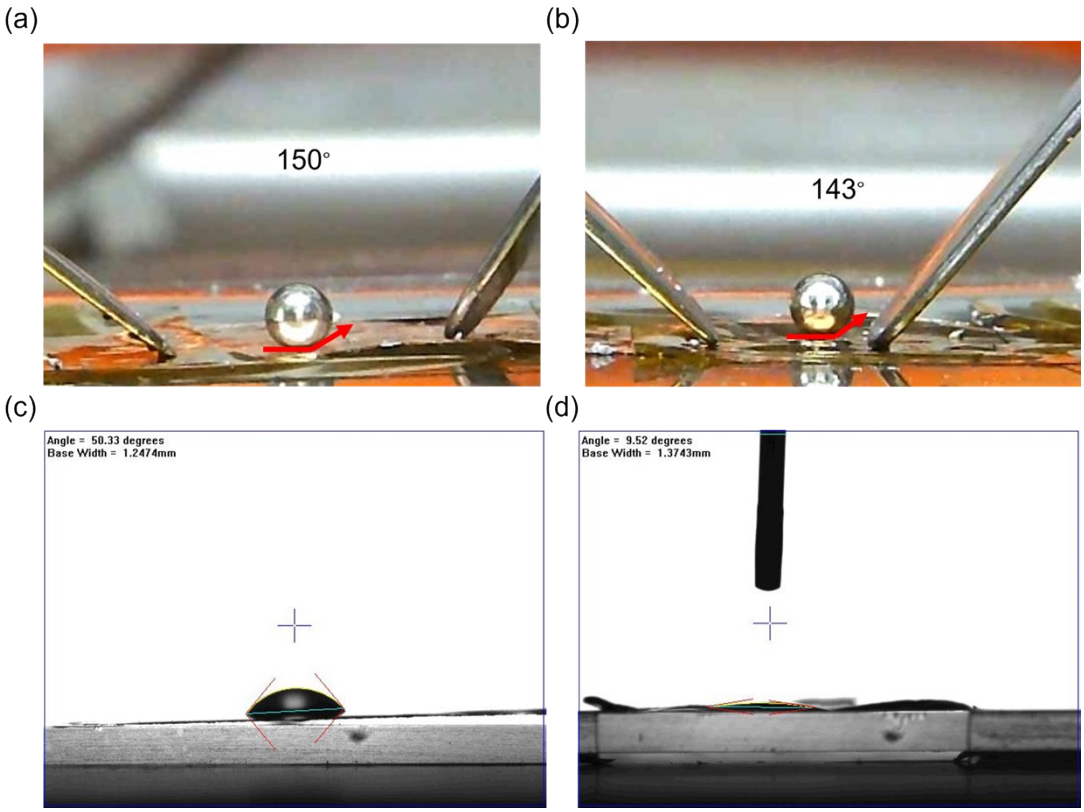

Fig. S2. The contact angle measurements for the (a, b) molten lithium at 300°C and (c, d) the organic electrolyte of 1 M LiTFSI + 2wt.% $LiNO_3$ in DOL/DME (1/1 vol.%) on (a, c) bare Cu and (b, d) G-Cu foils.

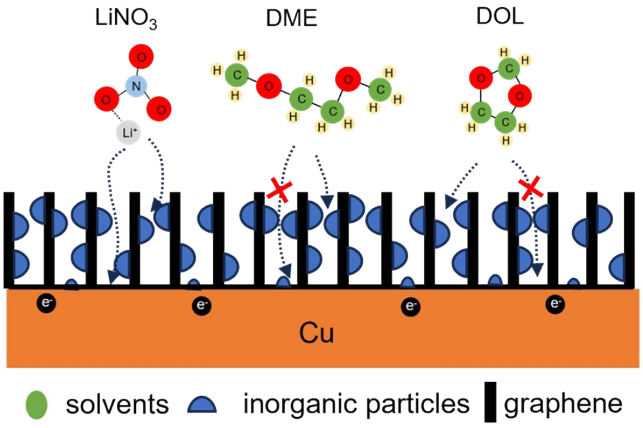

Fig. S3. The schematic diagram shows that the inorganic particles on the vertically aligned graphene layers impede the diffusion of solvent molecules to the Cu substrate for forming organic components in the SEI.

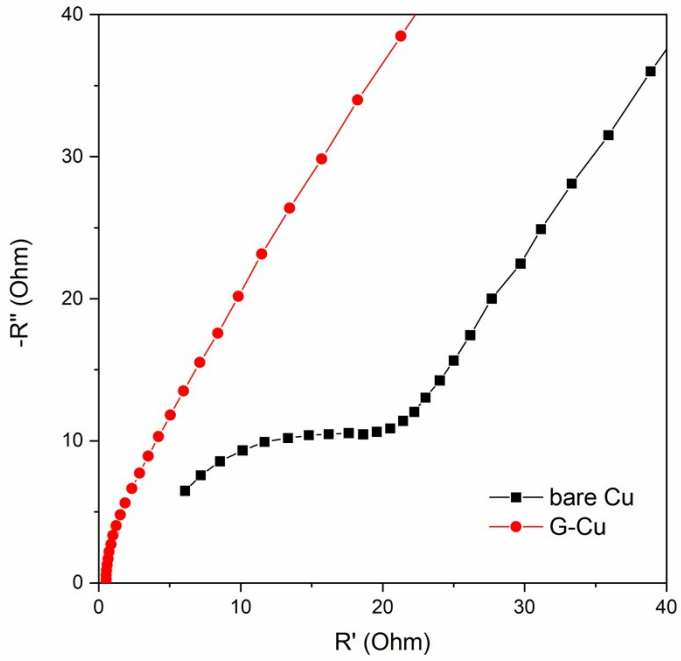

Fig. S4. Nyquist plots of bare Cu and G-Cu foils after a CV scan at 1 mV s^{-1} between 3.0 and 0.2 V (vs. Li^+/Li).

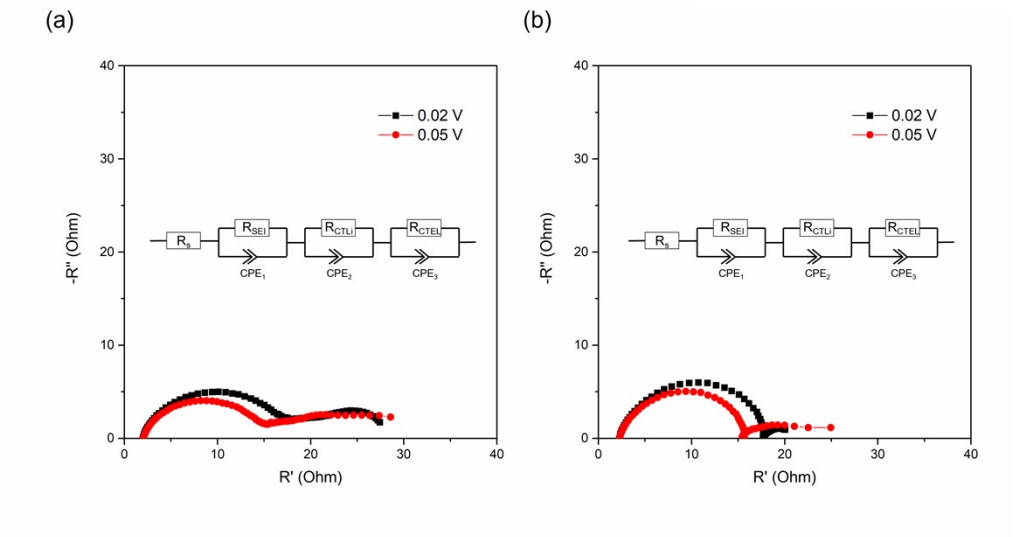

Fig. S5 Nyquist plots of the lithium deposits stripped for 15 min on (a) bare Cu and (b) G-Cu, measured at 0.02 and 0.05 V (vs. Li^+/Li) under a three-electrode mode.

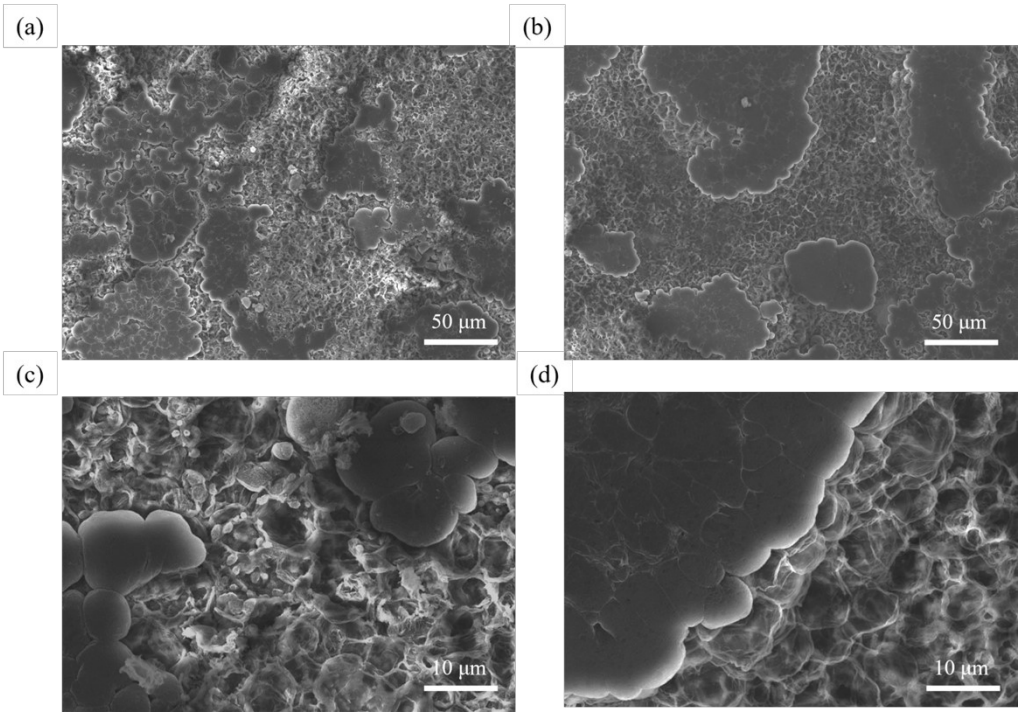

Fig. S6. SEM (a, b) regular and (c, d) enlarged top-view images for the lithium deposits electroplated at 1 mA cm^{-2} with an area capacity of 1 mAh cm^{-2} after the GCD test for 80 cycles on (a, c) bare and (b, d) G-Cu.

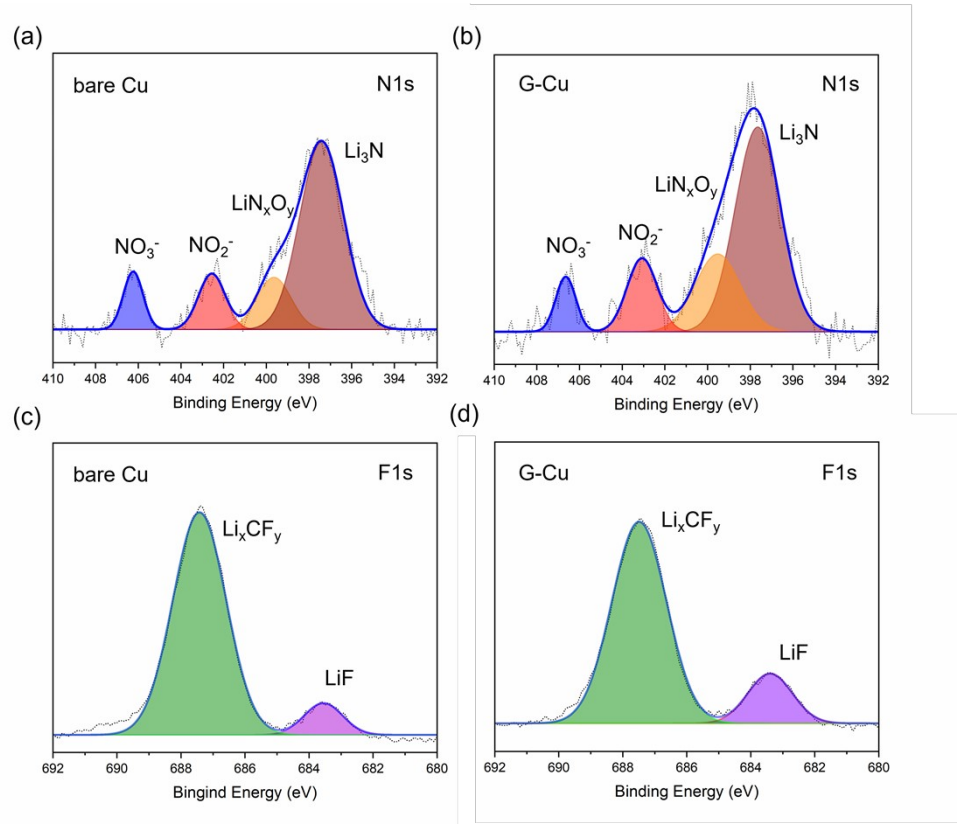

Fig. S7. (a, b) N1s and (c, d) F1s XPS spectra of the SEI layers on (a, c) bare Cu and (b, d) G-Cu after 20th GCD cycle at the fully discharged state in ZELMBs.

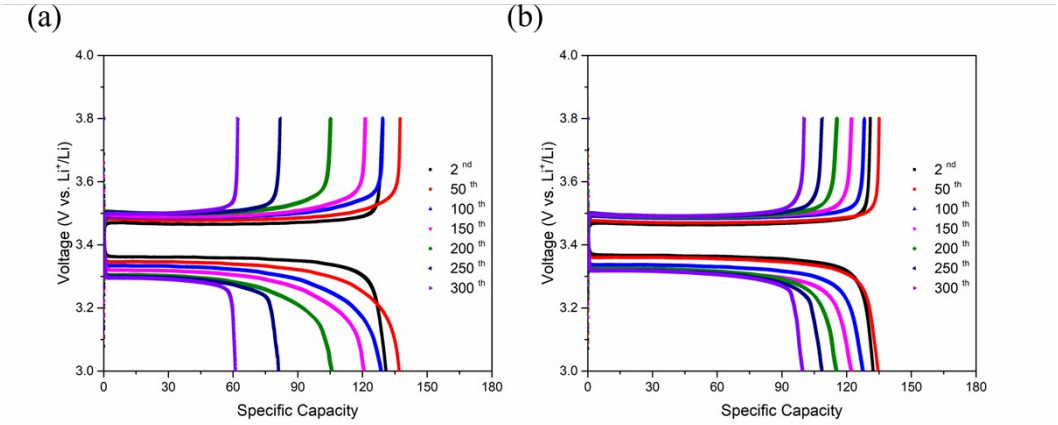

Fig. S8. GCD voltage profiles of the Li@Cu//LFP LMBs utilizing (a) bare Cu and (b) G-Cu.

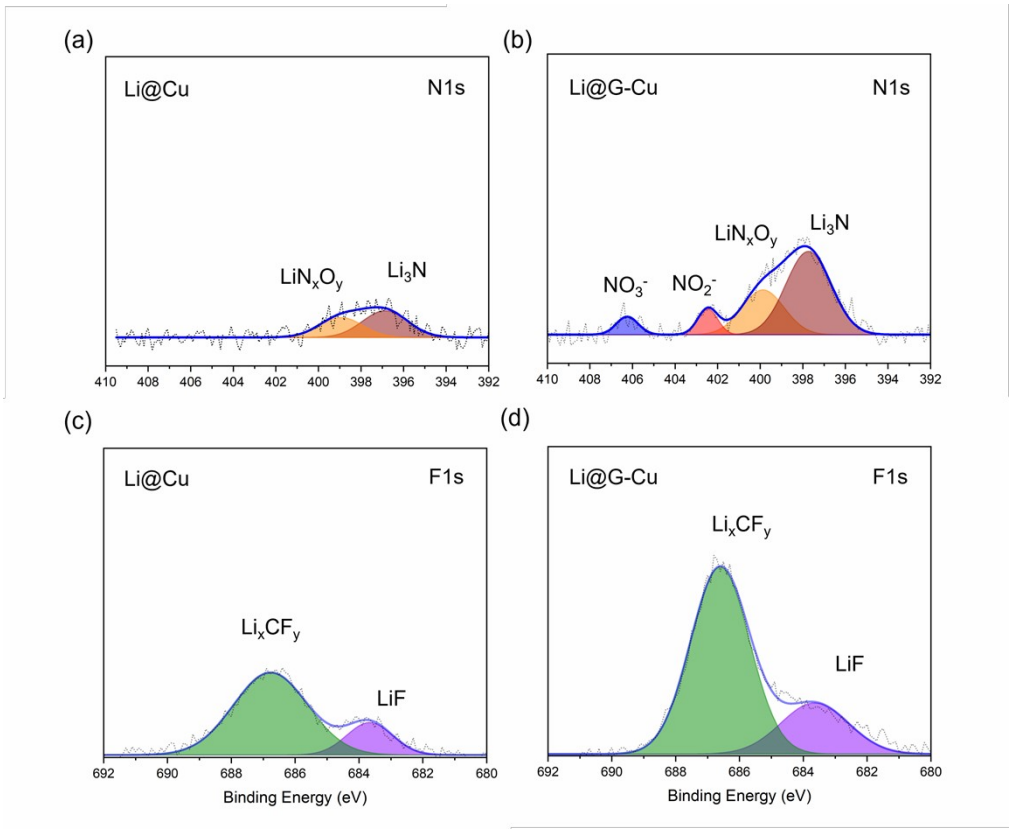

Fig. S9. (a, b) N1s and (c, d) F1s XPS spectra of the SEI adhered to the metallic lithium reservoir on (a, c) Li@Cu and (b, d) Li@G-Cu after the 20th GCD cycle in the LMBs with N/P=1.