Supporting Information

Establishing a multifunctional solid electrolyte interphase on a 3D host by ultra-fast double coating strategy for stable lithium metal batteries

Ji Young Maeng,[a,†] Minjun Bae,[a,†] Yonghwan Kim,[a] Dohyeong Kim,[a] Yujin Chang, [a] Seungman Park,[a] Juhyung Choi,[b] Eunji Lee,[a] Jeongyeon Lee,[d,] Yuanzhe Piao[a,b,c]**

[a] Department of Applied Bioengineering, Graduate School of Convergence Science and Technology, Seoul National University, 145 Gwanggyo-ro, Yeongtong-gu, Suwon-si, Gyeonggi-do, 16229, Republic of Korea

[b] Department of Transdisciplinary Studies, Graduate School of Convergence Science and Technology, Seoul National University, 145 Gwanggyo-ro, Yeongtong-gu, Suwon-si, Gyeonggi-do, 16229, Republic of Korea

[c] Advanced Institutes of Convergence Technology, 145 Gwanggyo-ro, Yeongtong-gu, Suwon-si, Gyeonggi-do, 16229, Republic of Korea

[d] School of Fashion and Textiles, The Hong Kong Polytechnic University, Hung Hom Kowloon, Hong Kong SAR, China

Corresponding author

* Yuanzhe Piao: Tel.: +82 31 888 9141; Fax: +82 31 888 9148; E-mail: parkat9@snu.ac.kr

* Jeongyeon Lee. E-mail: jaden-jy.lee@polyu.edu.hk

Author Contributions

† These authors equally contributed to this work

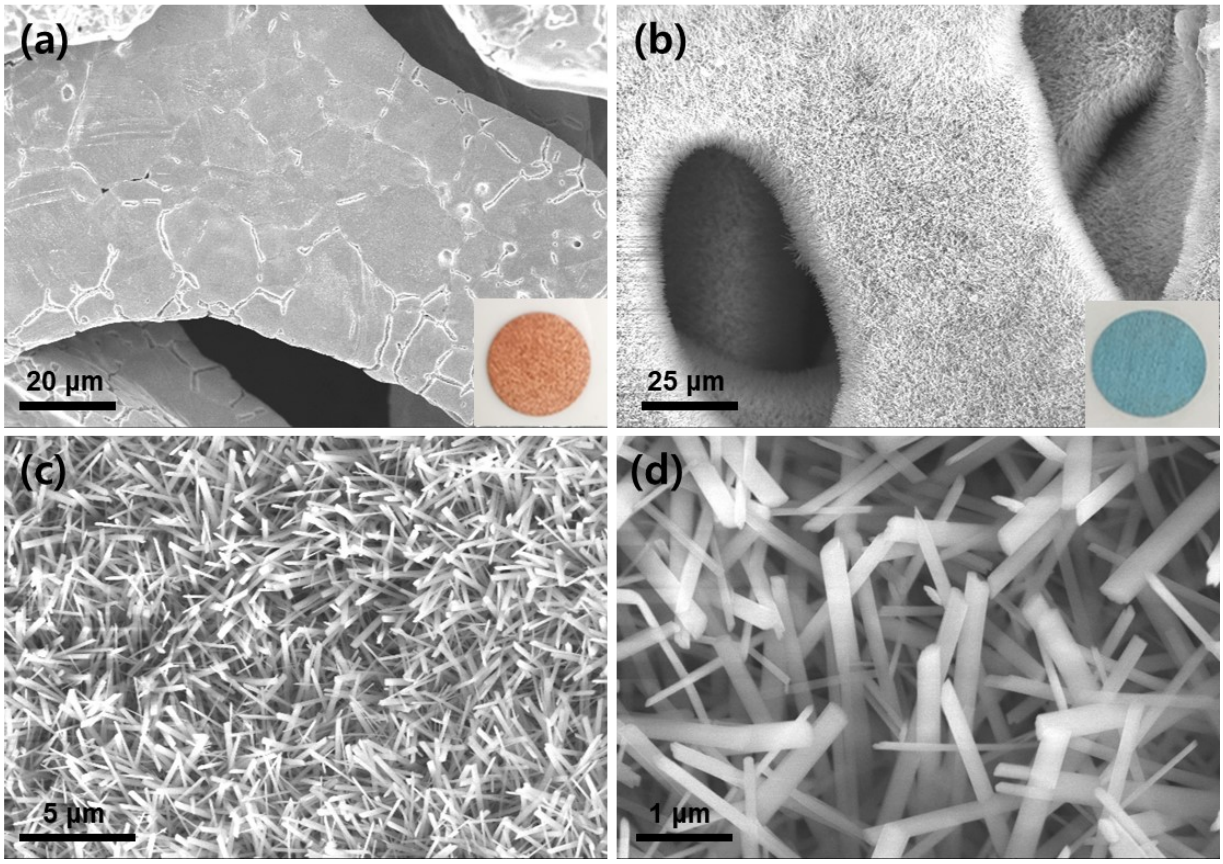

Fig. S1 SEM images of (a) pristine CF and (b-d) $Cu(OH)_2$@CF.

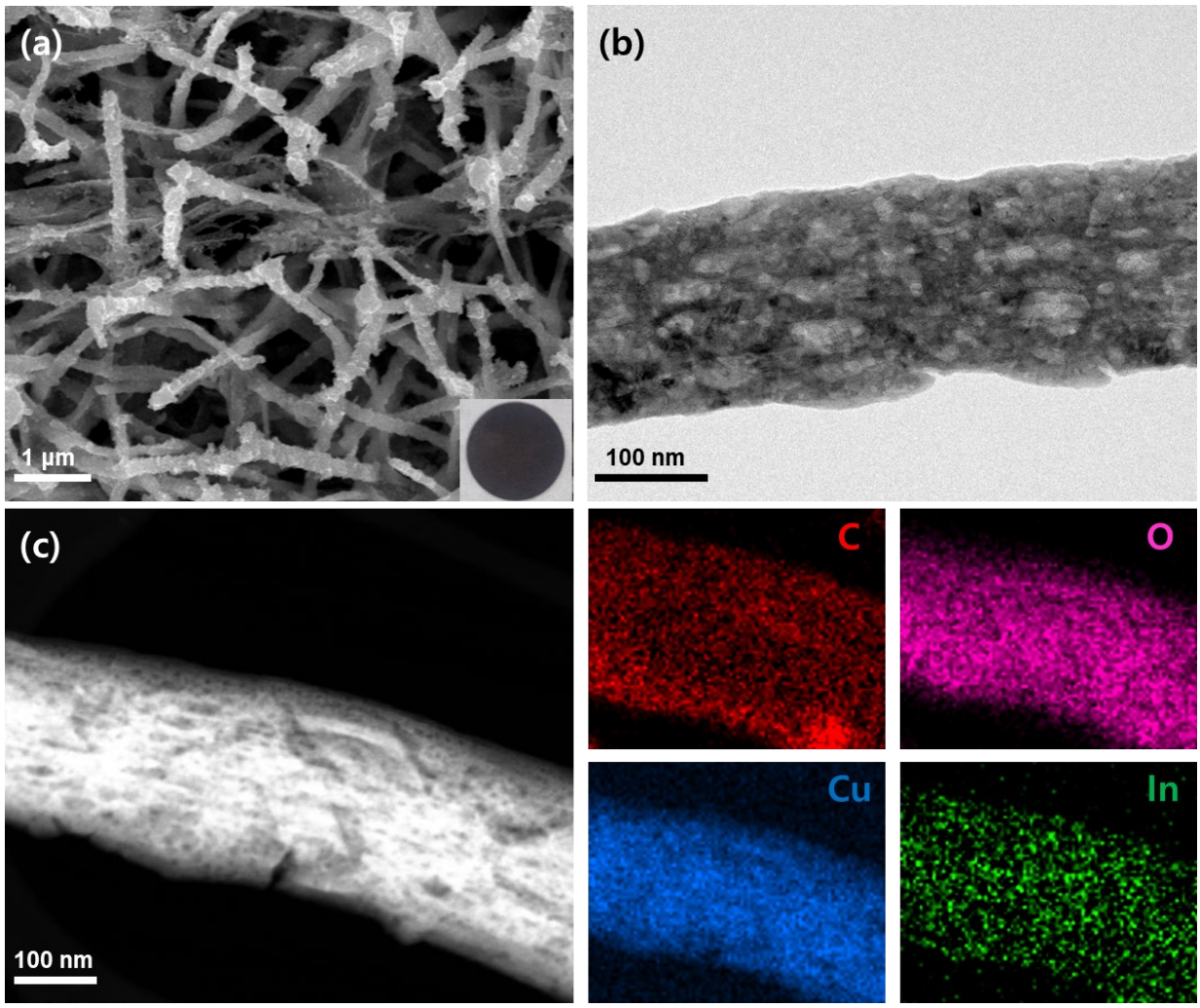

Fig. S2 (a) SEM, (b) TEM and (c) STEM-HADDF and corresponding EDS images of In@Cu.

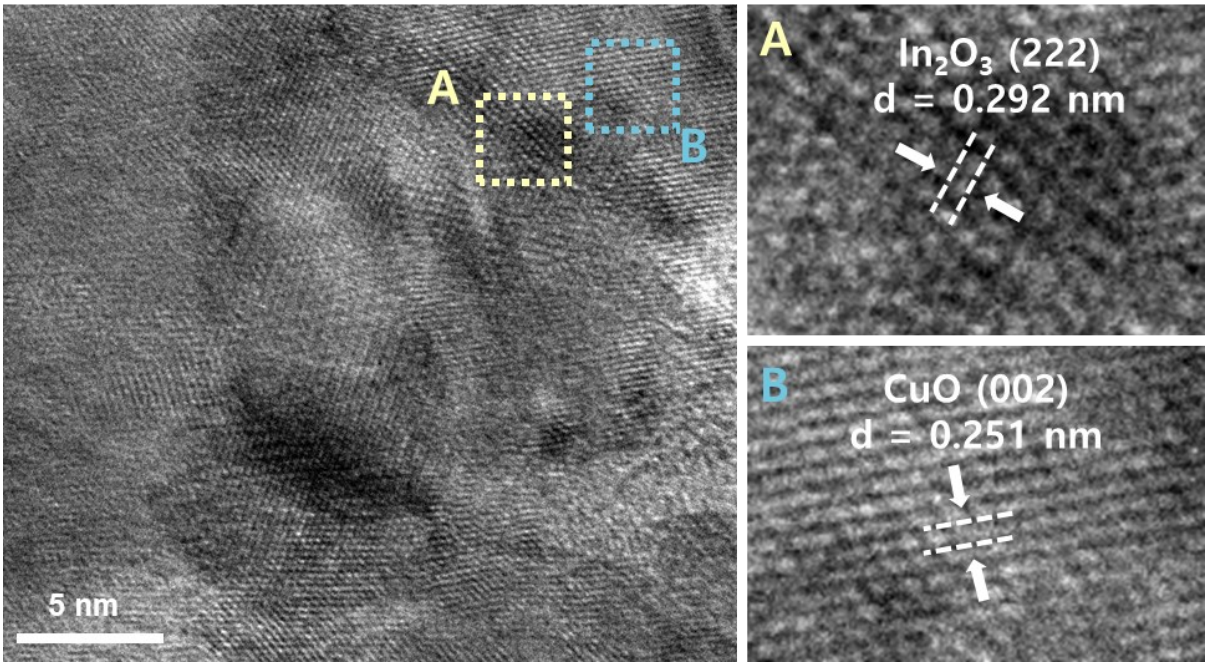

Fig. S3 HR-TEM images of In@Cu.

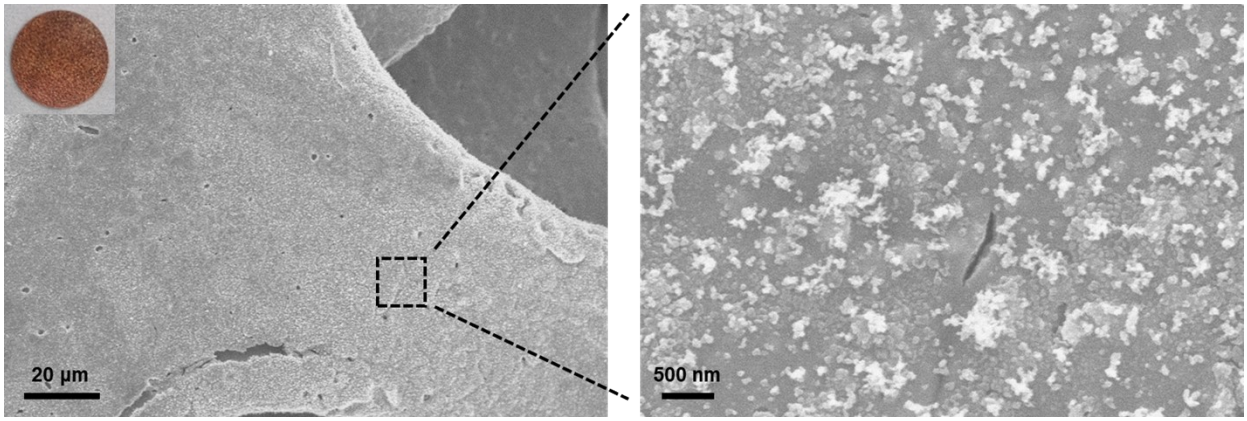

Fig. S4 SEM images of pristine CF after the flame treatment.

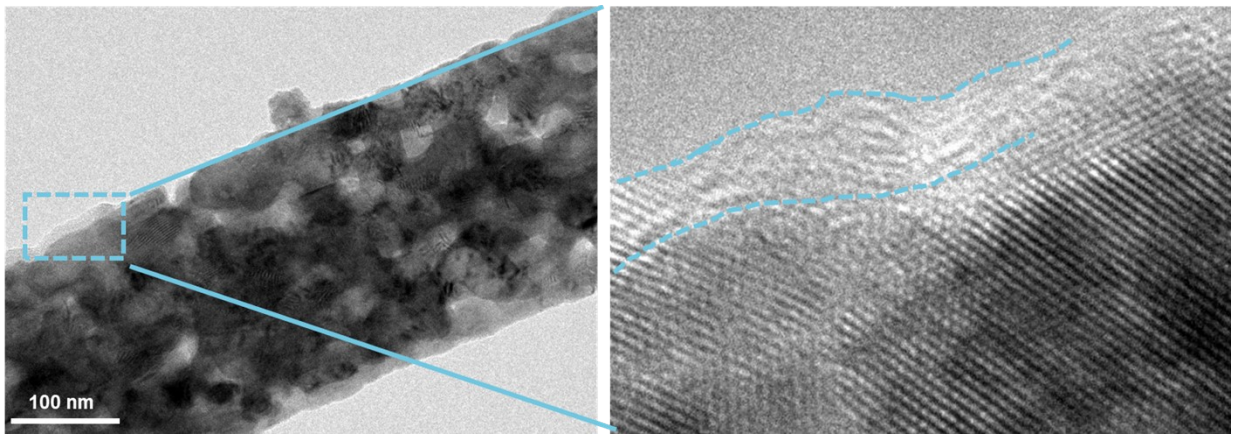

Fig. S5 TEM images of MSEI@Cu.

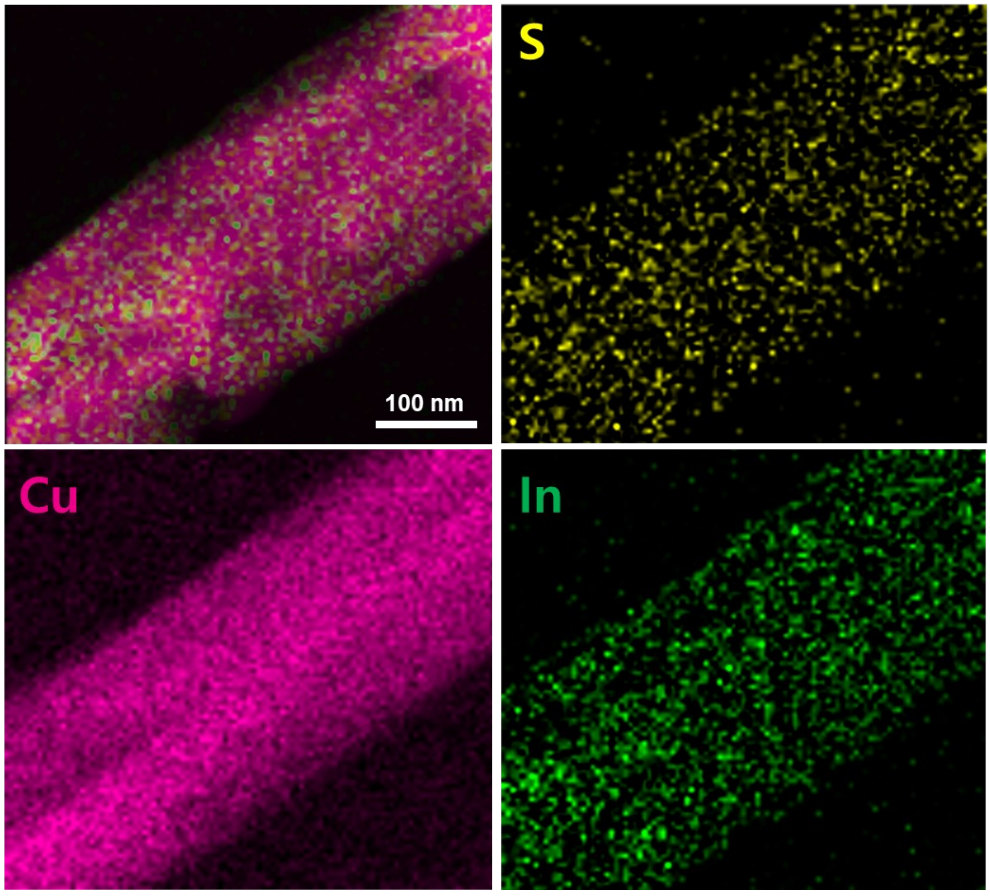

Fig. S6 The HADDF-STEM and corresponding EDS mapping images of MSEI@Cu.

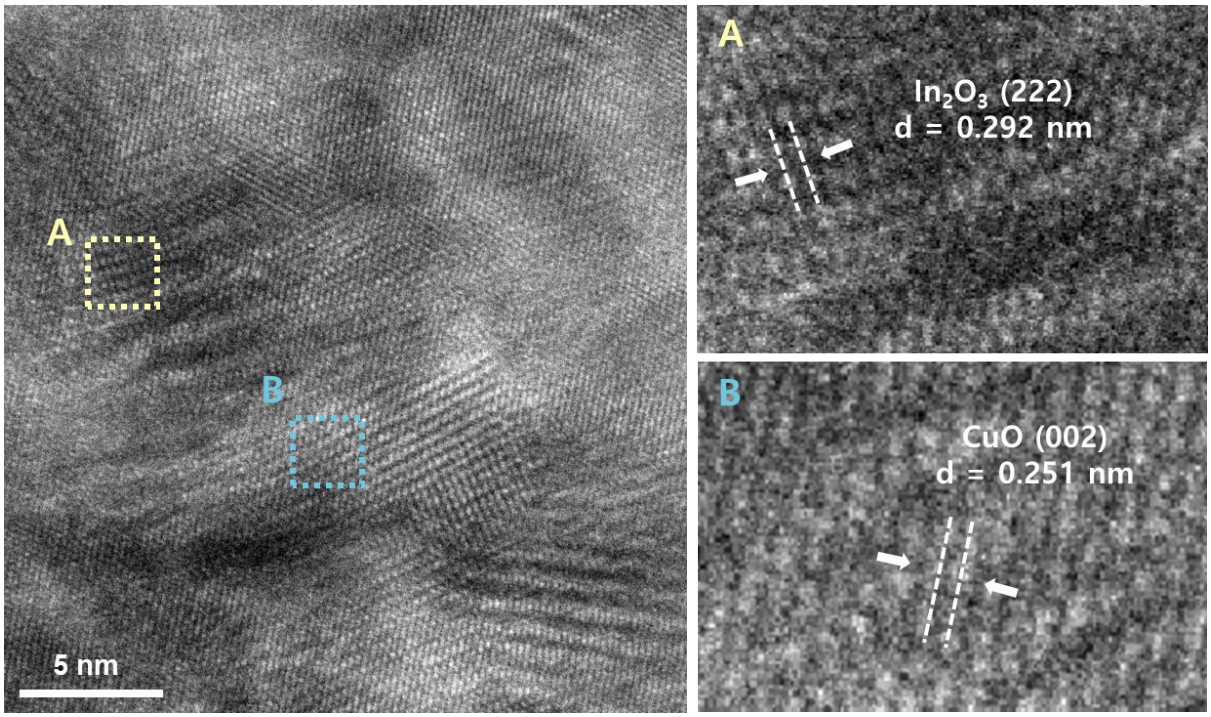

Fig. S7 HR-TEM images of MSEI@Cu.

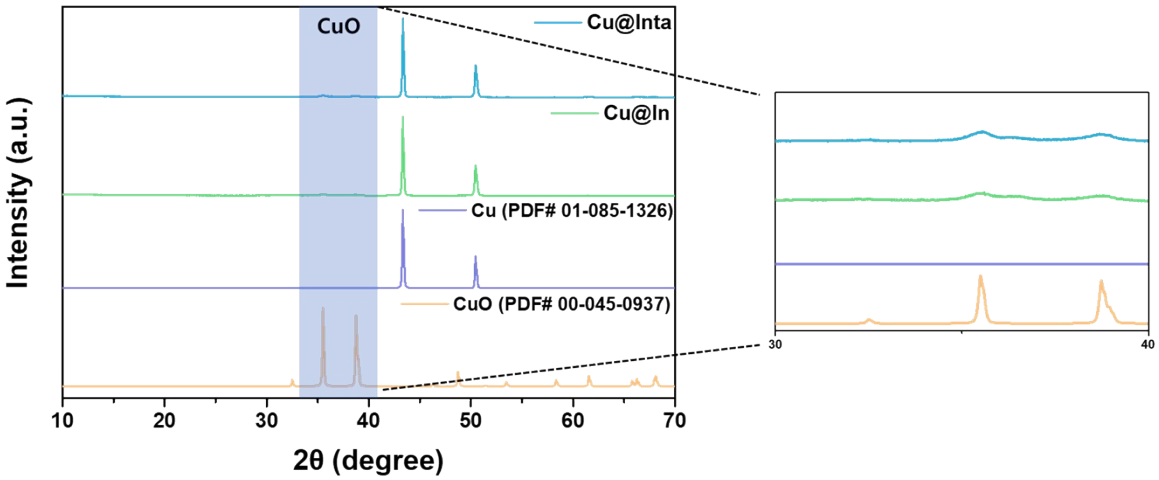

Fig. S8 XRD patterns of MSEI@Cu and In@Cu.

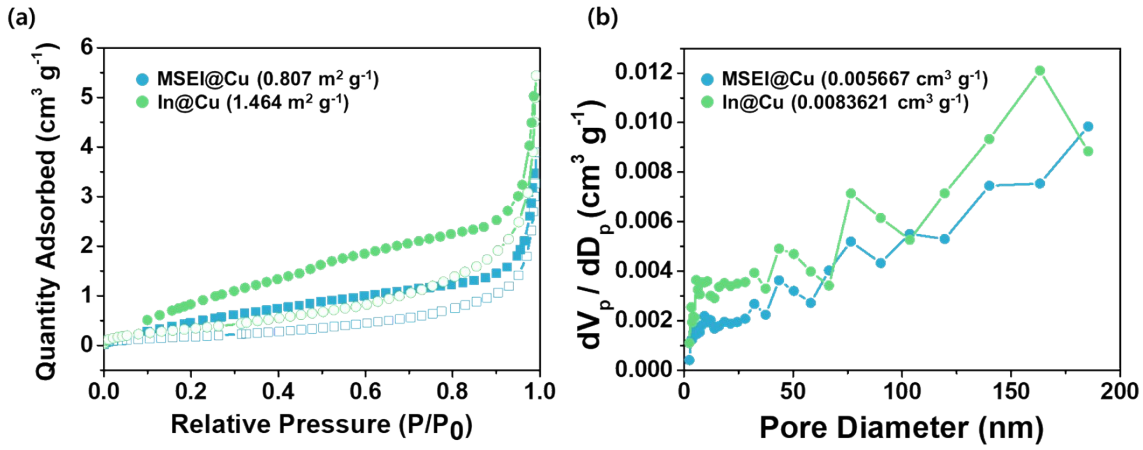

Fig. S9 (a) N_2 adsorption/desorption isotherms and (b) pore distributions of MSEI@Cu and In@Cu.

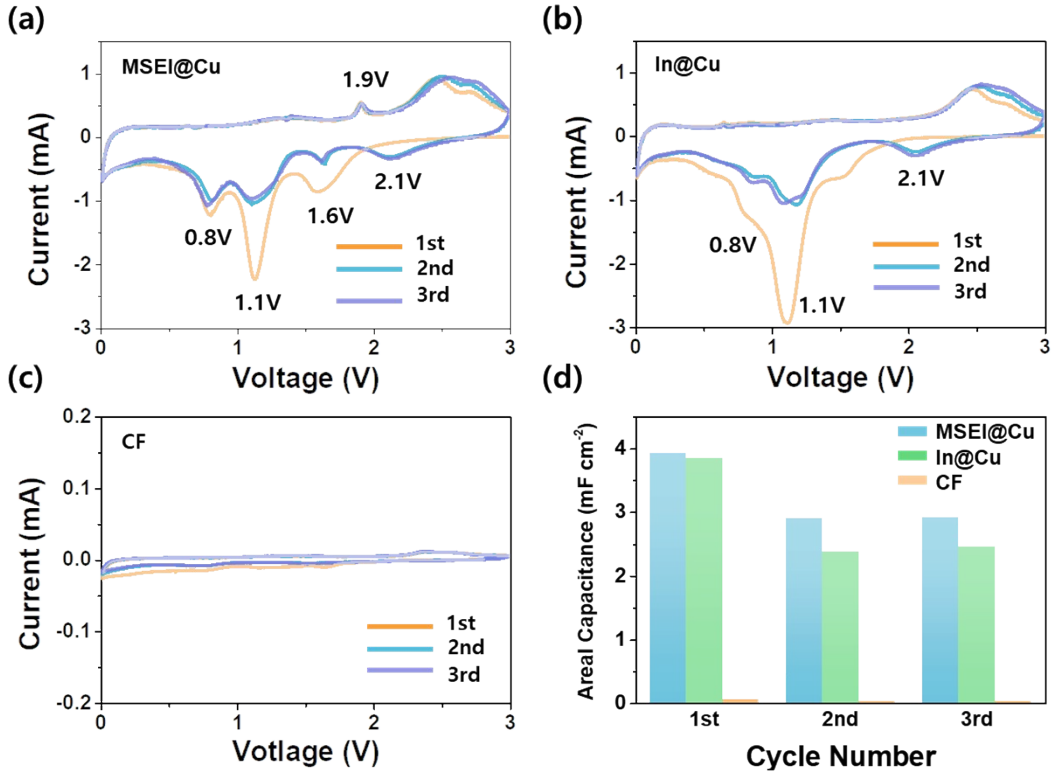

Fig. S10 The CV profiles of (a) MSEI@Cu, (b) In@Cu and (c) CF at a scanning rate of 0.1 mV s^{-1} between 0 and 3 V, and (d) the areal capacitance of each electrode measured at different cycle.

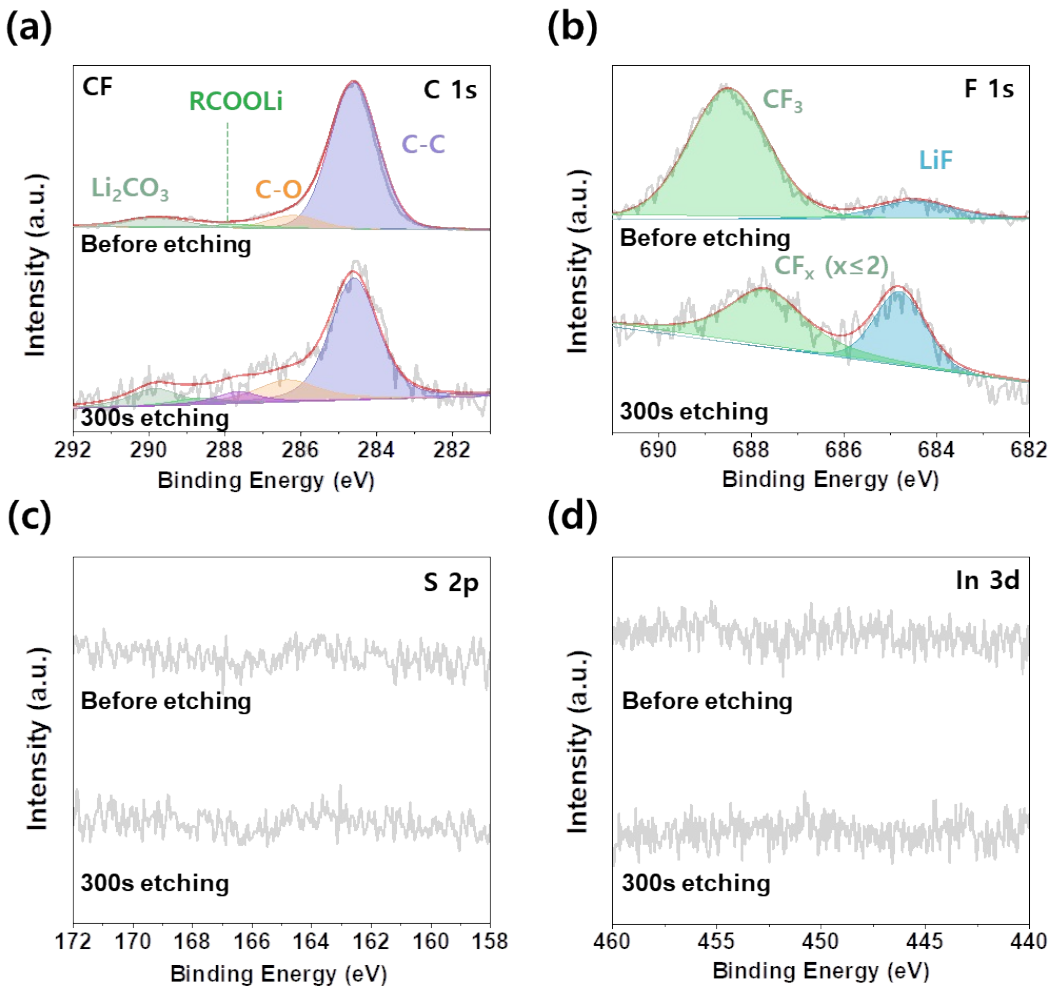

Fig. S11 The high-resolution (a) C 1s, (b) F 1s, (c) S 2p and (d) In 3d XPS spectra of CF after electrochemical SEI formation.

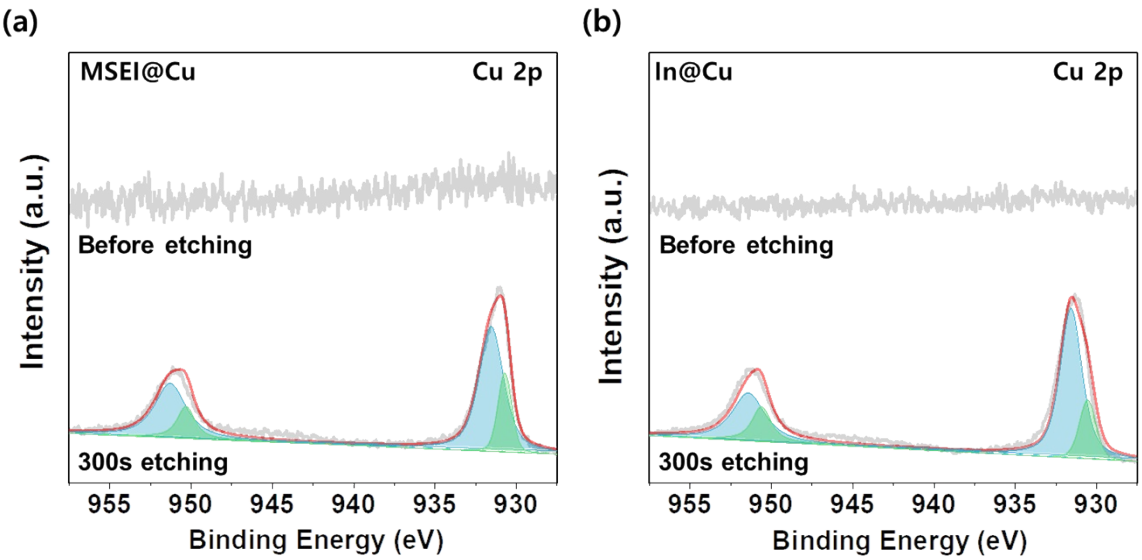

Fig. S12 The high-resolution Cu 2p XPS spectra of (a) MSEI@Cu and (b) In@Cu after electrochemical SEI formation.

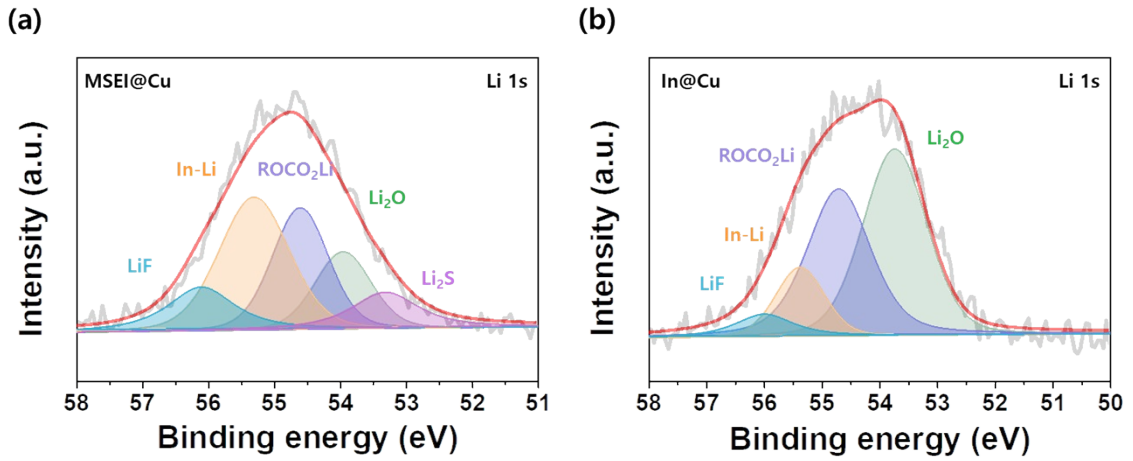

Fig. S13 The high-resolution Li 1s XPS spectra of (a) MSEI@Cu and (b) In@Cu after electrochemical SEI formation.

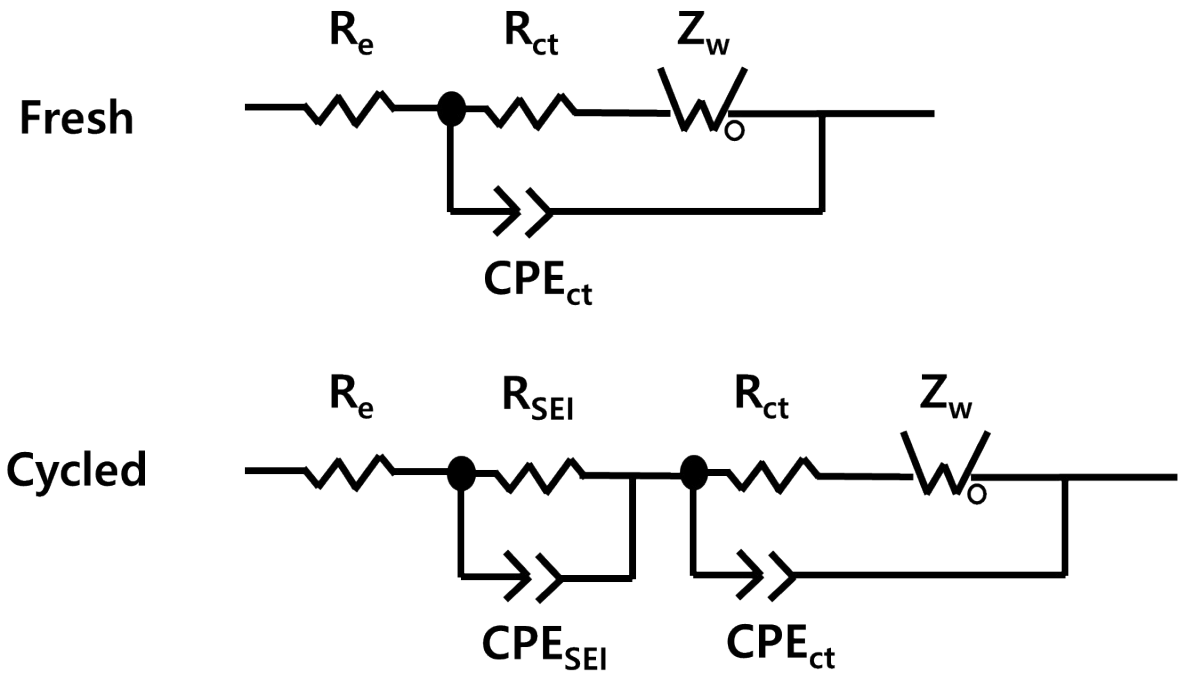

Fig. S14 EIS Equivalent circuit models.

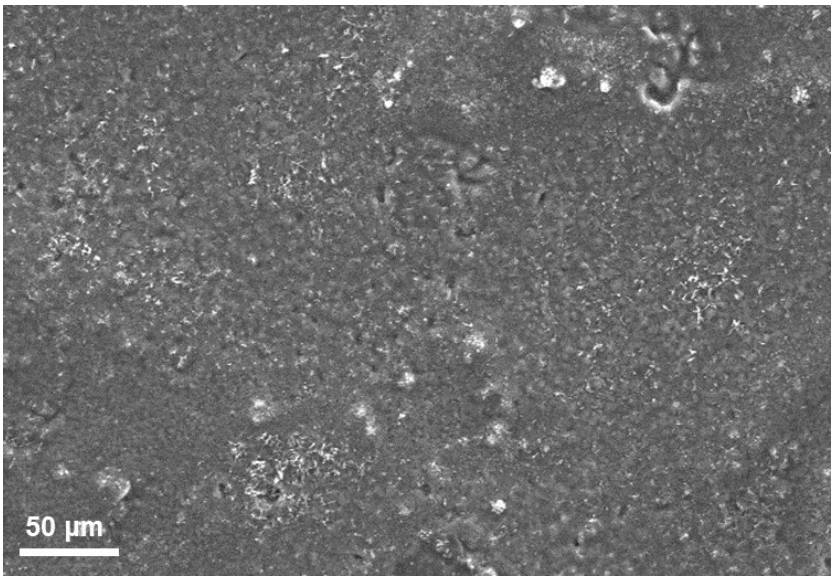

Fig. S15 The top SEM image of Li−MSEI@Cu.

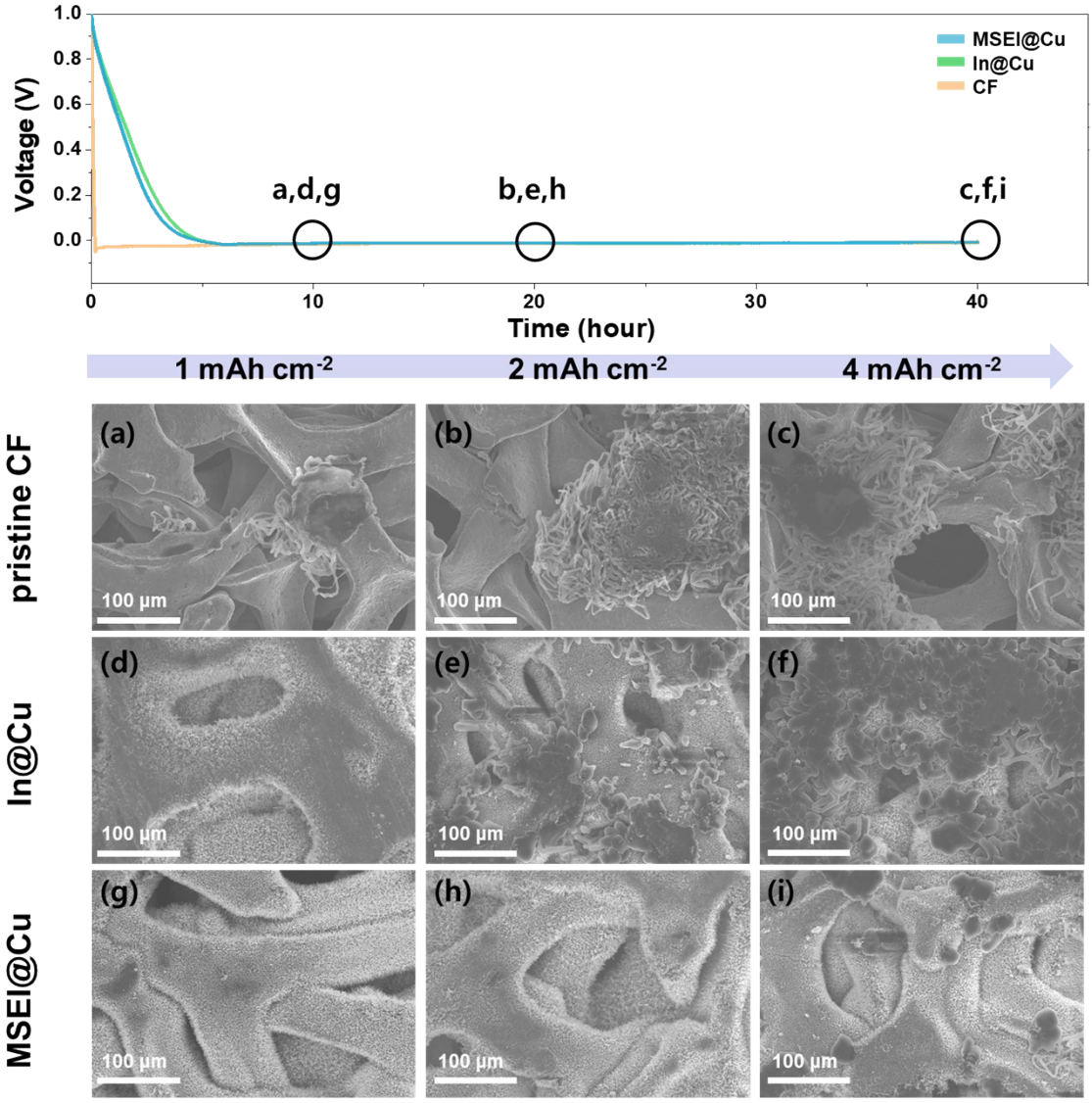

Fig. S16 SEM images (a–c) pristine CF, (d–f) In@Cu and (g–i) MSEI@Cu electrodes after Li electrodeposition at 0.1 mA cm^{-2}: (a, d, g) 1 mAh cm^{-2}, (b, e, h) 2 mAh cm^{-2}, (c, f, i) 4 mAh cm^{-2}.

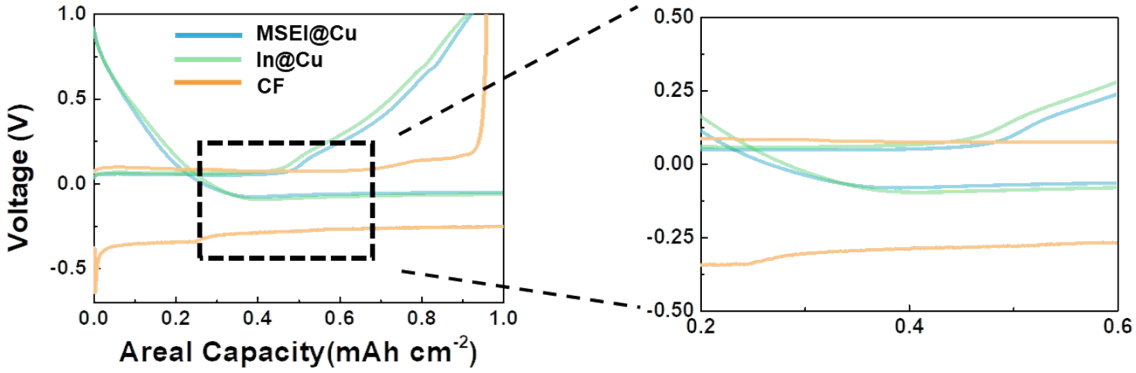

Fig. S17 The initial voltage profiles of MSEI@Cu, In@Cu and CF at 3 mA cm^{-2} and 1 mAh cm^{-2}.

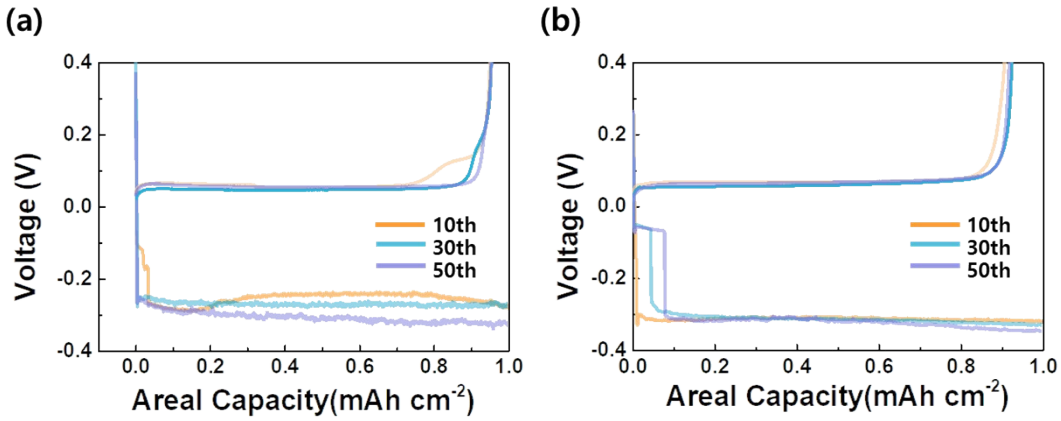

Fig. S18 Voltage profiles of CF at 10, 30 and 50th cycle at a current density of (a) 3 mA cm^{-2} and (b) 5 mA cm^{-2} with a fixed capacity of 1 mAh cm^{-2}.

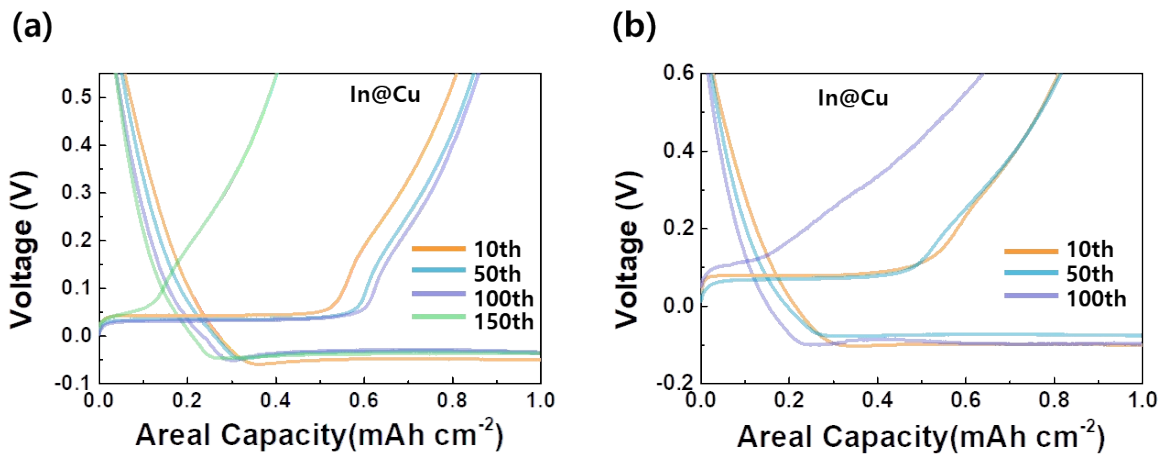

Fig. S19 Voltage profiles of In@Cu at 10, 50, 100 and 150th cycle at a current density of (a) 3 mA cm^{-2} and (b) those at 10, 50 and 100th cycle at a current density of 5 mA cm^{-2}. Both profiles operated under a fixed capacity of 1 mAh cm^{-2}.

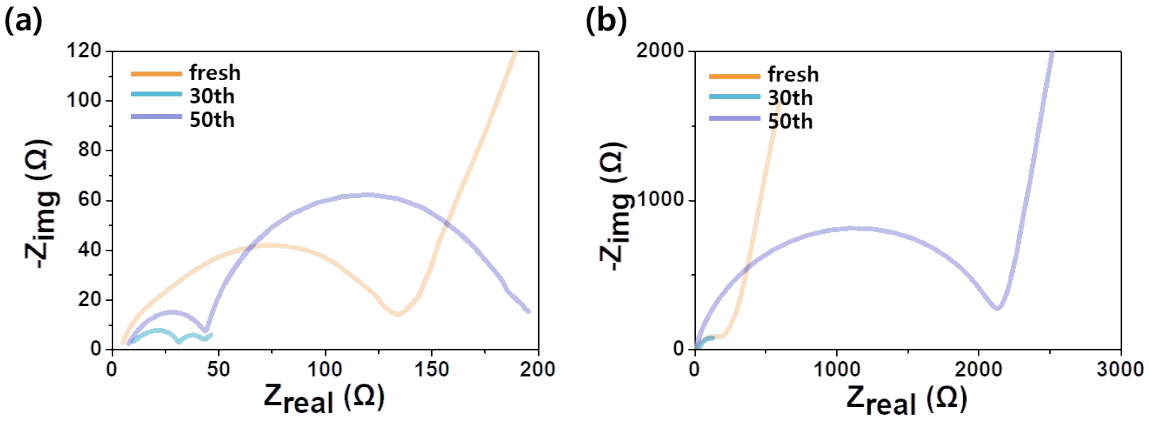

Fig. S20 Nyquist plots of (a) In@Cu (b) CF at fresh state, 30 and 50th cycle.

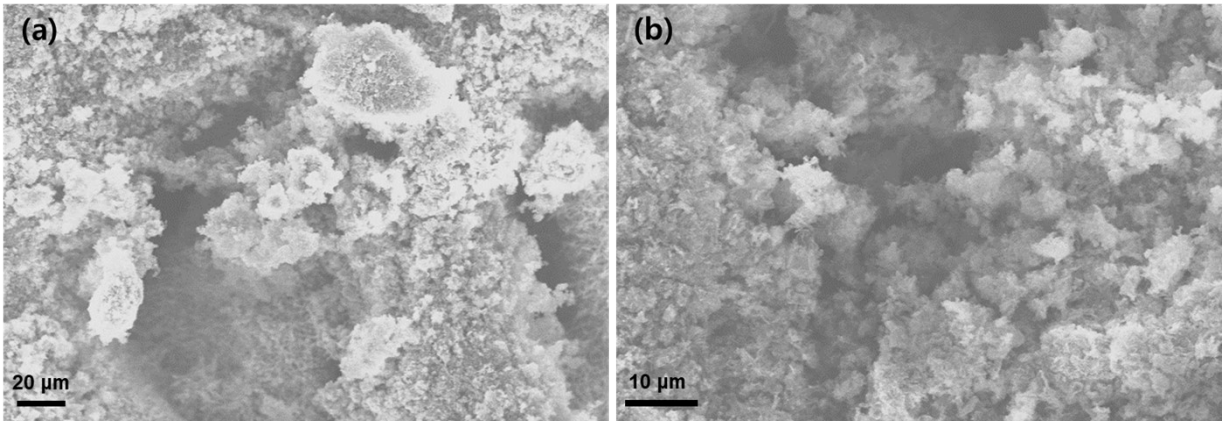

Fig. S21 The SEM images of In@Cu after 50 cycles at 3 mA cm^{-2} and 1 mAh cm^{-2}.

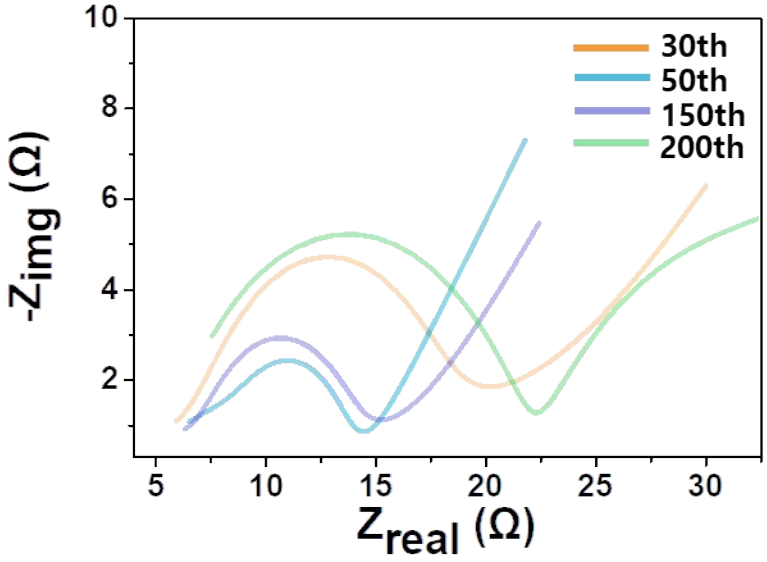

Fig. S22 Nyquist plots of bulk Li symmetric cells after 30, 50, 150, 200th cycle at a current density of 1 mA cm^{-2} and a capacity of 1 mAh cm^{-2}.

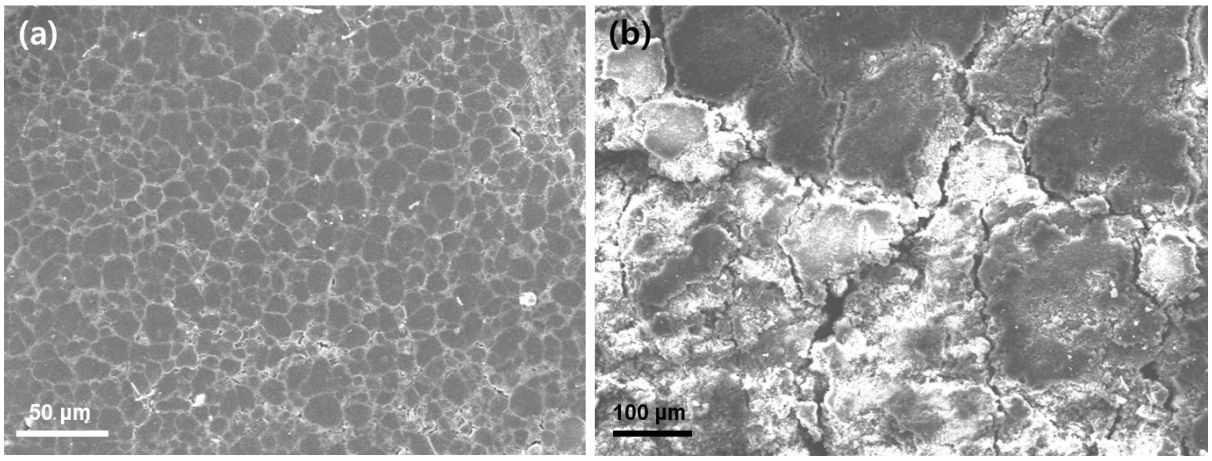

Fig. S23 Low-magnification SEM images of (a) Li−MSEI@Cu and (b) bulk Li electrodes after 200 cycles at 1 mA cm^{-2} and 1 mAh cm^{-2}.

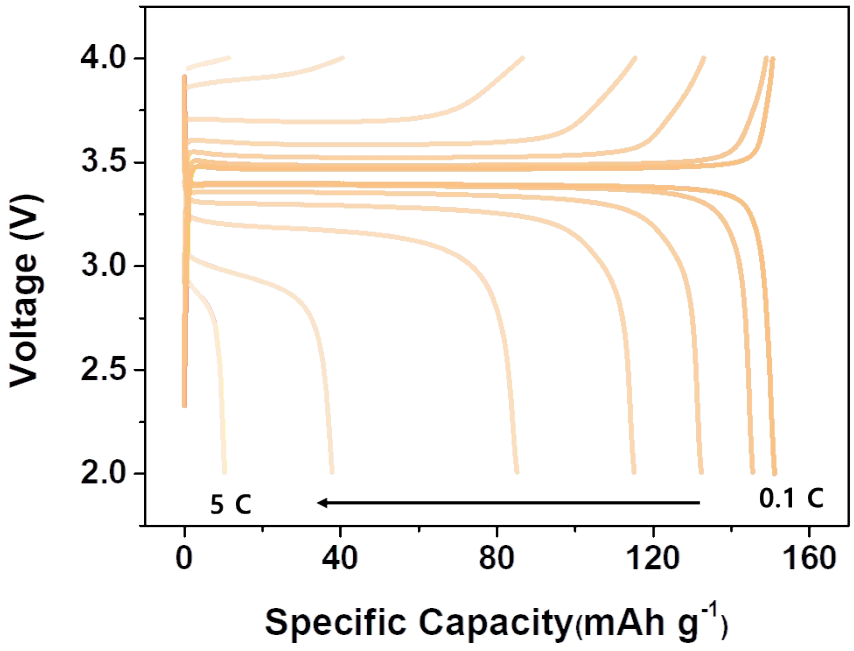

Fig. S24 Representative voltage profiles of the rate-performance of bulk Li|LFP full cell at various C-rates.

Name	MSEI@Cu (Atomic %)	In@Cu (Atomic %)
S2p	6.43	0
C1s	22.99	40.96
In3d	6.12	15.53
O1s	45.7	40.48
Cu2p	18.76	3.03

Table S1. XPS peak table of Cu@MSEI and In@Cu

Name	Li-MSEI@Cu Before Etching (Atomic %)	Li-In@Cu Before Etching (Atomic %)	Li-MSEI@Cu After Etching (Atomic %)	Li-In@Cu After Etching (Atomic %)
Li1s	32.93	26.76	45.02	45.19
S2p	1.64	0	1.93	0
C1s	27.73	37.62	11.1	18.76
In3d	0	0	1.77	1.08
O1s	33.64	31.84	34.54	28.74
F1s	3.07	2.83	2.99	1.88
Cu2p	0	0	2.13	4.35

Table S2. XPS peak table of Li-Cu@MSEI and Li-In@Cu

	Electrode name	$LiFePO_4$ loading mass (mg cm^{-2})	Cycle number	Reference
1	CuFePBA−150/ Cu	1.5	500	[S1]
	CuFePBA−150/ Cu	12.4	120	[S1]
2	3D Li−Zn@Cu	2	250	[S2]
3	LVCF@Li anodes	12	200	[S3]
4	Li_2O@CuNA/CF	5	500	[S4]
5	Li@Cu_xO−7	8	500	[S5]
	Li@Cu_xO−7	12	100	[S5]
6	$Ti_3C_2T_x$@Cu	2	225	[S6]
7	CuSnAl@Cu	3	300	[S7]
8	Cu−Ag alloy	2.8	500	[S8]
9	Cu_3Sn/CP	12	100	[S9]
10	3D porous ZnO−nVF@Cu	1	200	[S10]
11	Our work	6.5	500	
	Our work	13.5	200	

Table S3. Comparison of long-term cycling performance of LFP-based full cells between our work and other previously reported 3D Cu hosts.

References

[S1] Y. Cai, B. Qin, J. Lin, C. Li, X. Si, J. Cao, J. Qi, *ACS Appl. Mater. Interfaces*, 2021, **13**, 23803.

[S2] Y. Ye, Y. Liu, J. Wu, Y. Yang, *J. Power Sources*, **2020**, *472*, 228520.

[S3] T. Liu, S. Chen, W. Sun, L.P. Lv, F.H. Du, H. Liu, Y. Wang, *Adv. Funct. Mater.*, 2021, **31**, 2008514.

[S4] L. Tan, X. Li, M. Cheng, T. Liu, Z. Wang, H. Guo, G. Yan, L. Li, Y. Liu, J. Wang, *J. Power Sources*, 2020, **463**, 228178.

[S5] Y. Nie, X. Dai, J. Wang, Z. Qian, Z. Wang, H. Guo, G. Yan, D. Jiang, R. Wang, *J. Energy Chem.*, 2022, **75**, 285.

[S6] L. Zhang, Q. Jin, K. Zhao, X. Zhang, L. Wu, *ACS Appl. Energy Mater.*, 2022, **5**, 2514.

[S7] Y. Lv, Q. Zhang, C. Li, C. Ma, W. Guan, X. Liu, Y. Ding, *ACS Sustain-able Chem. Eng.*, 2022, **10**, 7188.

[S8] Y. Liu, S. Huang, Q. Meng, Y. Fan, B. Wang, Y. Yang, G. Cao, H. Zhang, *J Alloys Compd*, 2021, **885**, 160882.

[S9] J. Jiang, J. Lu, Y. Ou, G. Liu, S. Lu, Y. Jiang, B. Zhao, J. Zhang, *ACS Appl. Mater. Interfaces*, 2022, **14**, 2930.

[S10] G. Luo, X. Yin, D. Liu, A. Hussain, F. Liu, X. Cai, *ACS Appl. Mater. Interfaces*, 2022, **14**, 33400.