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Supporting Information

Dual-Regulation of Ion/Electron in 3D Cu-Cu_xO host to Guide Uniform Lithium

Growth for High-Performance Lithium Metal Anodes

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Fig. S1. a) SEM image of the $Cu-Cu(OH)_2$. b-c) SEM images of solid $Cu(OH)_2$ nanowires with different magnification.



Fig. S2. TEM images of porous Cu_xO nanowires with different magnification.



Fig. S3. HR-TEM images of Cu_xO nanowires.



Fig. S4. Initial discharge profile of the Cu-Cu_xO and Cu foam.



Fig. S5. SEM images of the Cu-Cu_xO after the lithiation reaction.



Fig. S6. XRD pattern of the Cu-Cu_xO host after cycling.



Fig. S7. a-b) SEM images of Cu foam after plating 0.05 mAh cm⁻² Li. c-d) SEM images of the Cu-Cu_xO after plating 0.05 mAh cm⁻² Li.



Fig. S8. SEM images of the Cu-Cu_xO after plating 10 mAh cm⁻² Li.



Fig. S9. SEM images of the a) top surface and b) bottom surface of the Cu-Cu_xO after plating 3 mAh cm⁻² Li. SEM images of the c) top surface and d) bottom surface of Cu foam after plating 3 mAh cm⁻² Li (Inset is the corresponding optical photo). SEM images of the e) top surface and f) bottom surface of the Cu-Cu_xO after plating 5 mAh cm⁻² Li. SEM images of the g) top surface and h) bottom surface of Cu foam after plating 5 mAh cm⁻² Li (Inset is the corresponding optical photo).



Fig. S10. Cross-section SEM images of the Cu-Cu_xO after plating a) 1 mAh cm⁻², b) 3 mAh cm⁻² and c) 5 mAh cm⁻² Li.



Fig. S11. a-c) SEM images of Cu foam after 50 cycles with different magnification. d-f) SEM images of the Cu-Cu_xO after 50 cycles with different magnification.



Fig. S12. (a-c) SEM images of the 3D Cu-Cu_xO host after cycling. (d) EDS mapping images of the nanowires after cycling.



Fig. S13. TEM images of the nanowires after cycling with different magnification.



Fig. S14. Schematic illustration of the in-situ optical microscopy observation device.



Fig. S15. Long-term CE of the Cu-Cu_xO host with areal capacity of 1 mAh cm⁻² at 1 mA cm⁻².



Fig. S16. CE of Li plating/stripping with areal capacity of 3 mAh cm⁻² at 1 mA cm⁻².



Fig. S17. CE of Li plating/stripping with a) areal capacity of 2 mAh cm⁻² at 5 mA cm⁻² and b) areal capacity of 5 mAh cm⁻² at 5 mA cm⁻².



Fig. S18. a) Voltage profile of Li plating/stripping on the Cu-Cu_xO with areal capacity of 3 mAh cm⁻² at 1 mA cm⁻². b) Voltage profile of Li plating/stripping on the Cu-Cu_xO with areal capacity of 1 mAh cm⁻² at 3 mA cm⁻² and 5 mA cm⁻².



Fig. S19. The EIS profile of Cu foam and the Cu-Cu_xO host. a) Freshly assembled coin cell before lithiation. b) Coin cell after lithiation. c) Equivalent circuit diagram to simulate Nyquist plots of Cu-Cu_xO host.



Fig. S20. Voltage profiles of Li plating/stripping with areal capacity of 3 mAh cm⁻² at 3 mA cm⁻².



Fig. S21. Long-term cycling performance of Cu-Cu_xO@Li anode.

Table S1. Comparison of CE between 3D Cu-Cu_xO and other metal-based current collectors in the literatures.

Current Collector	Current Density	Capacity	Cycle Number	CE (%)	Ref.
Cu Nanowires (Cu NWs)	1 mA cm^{-2}	2 mAh cm^{-2}	200	98.6	1
	2 mA cm^{-2}	2 mAh cm^{-2}	50	97.6	
	5 mA cm^{-2}	2 mAh cm^{-2}	50	97.1	
Chemical Dealloying 3D	0.5 mA cm^{-2}	1 mAh cm^{-2}	240	97	2
Porous Cu (3D-Cu)	1 mA cm^{-2}	1 mAh cm^{-2}	140	97	
Carbon Modified Ni Foam (CMN)	0.5 mA cm^{-2}	2 mAh cm^{-2}	475	98	3
O-CNT Modified Ni Foam (O- CNT)	1 mA cm^{-2}	2 mAh cm^{-2}	200	99	4
LCNM@Ni	2 mA cm^{-2}	4 mAh cm^{-2}	200	99	5
Cu ₉₉ Zn Foils (Cu ₉₉ Zn)	0.2 mA cm^{-2}	0.4 mAh cm^{-2}	400	99	- 6
	0.5 mA cm^{-2}	1 mAh cm^{-2}	200	97	
Vertical Graphene Nanowalls Modified Ni Foam (VGN)	0.5 mA cm^{-2}	1 mAh cm ⁻²	250	97	7
CNT Sponge on Cu Foil (C- host@Cu)	1 mA cm^{-2}	3 mAh cm^{-2}	75	96	8
3D porous Cu skeleton (Cu	0.5 mA cm^{-2}	1 mAh cm^{-2}	700	95	9
skeleton)	1 mA cm^{-2}	1 mAh cm^{-2}	400	95	
Lithiophilic Cu-CuO-Ni Hybrid Structure (Cu-CuO-Ni)	1 mA cm^{-2}	1 mAh cm ⁻²	200	95	10
Graphene Coating on Cu Foam	0.5 mA cm^{-2}	1 mAh cm^{-2}	250	98.6	11
	2 mA cm^{-2}	1 mAh cm^{-2}	150	97.4	
(GN@Cu)	5 mA cm^{-2}	1 mAh cm^{-2}	90	96.5	
VA-CuO on Cu Foil (VA-	0.5 mA cm^{-2}	1 mAh cm^{-2}	180	94	12
CuO-Cu)	1 mA cm^{-2}	1 mAh cm^{-2}	180	94	
	0.5 mA cm^{-2}	1 mAh cm^{-2}	150	97.5	13
N-doped Graphene Modified	1 mA cm^{-2}	2 mAh cm^{-2}	50	97	
3D Porous Cu (3DCu@NG)	0.5 mA cm^{-2}	2 mAh cm^{-2}	100	97.6	
	0.5 mA cm^{-2}	4 mAh cm^{-2}	100	97.8	
Compact 3D Cu (3D Cu)	1 mA cm^{-2}	1 mAh cm^{-2}	200	97.9	14
Zn/Cu _{0.7} Zn _{0.3} /CF (Cu-Zn)	1.5 mA cm^{-2}	1 mAh cm^{-2}	300	98	15
	2.5 mA cm^{-2}	1.5 mAh cm^{-2}	150	97	
	5 mA cm^{-2}	3 mAh cm^{-2}	50	96	
CuO@Ti-mesh (CTM)	1 mA cm^{-2}	1 mAh cm^{-2}	500	97.3	- 16
	5 mA cm^{-2}	1 mAh cm^{-2}	100	95	
Graphene on Cu Mesh (VGCM)	2 mA cm ⁻²	1 mAh cm ⁻²	250	97	17
	3 mA cm^{-2}	1 mAh cm ⁻²	200	97	
	5 mA cm^{-2}	1 mAh cm ⁻²	150	97	
	1 mA cm^{-2}	5 mAh cm^{-2}	80	98	

Cu ₂ S NWs on Cu Foam (Cu ₂ S/Cu)	0.5 mA cm^{-2}	1 mAh cm^{-2}	150	98.9	
	1 mA cm^{-2}	1 mAh cm^{-2}	150	95.5	18
	3 mA cm^{-2}	1 mAh cm^{-2}	100	92	
Cu@NPCN	1 mA cm^{-2}	1 mAh cm^{-2}	400	99	10
	3 mA cm^{-2}	1 mAh cm^{-2}	250	98.2	19
Deposition-regulating Scaffold	1 mA cm^{-2}	1 mAh cm^{-2}	500	98.1	20
(DRS)	0.5 mA cm^{-2}	2 mAh cm^{-2}	350	97	
3D Porous Cu (P-Cu)	2 mA cm^{-2}	2 mAh cm^{-2}	200	97.6	
	2 mA cm^{-2}	4 mAh cm^{-2}	62	97.1	21
	1 mA cm^{-2}	3 mAh cm^{-2}	385	98	
	3 mA cm^{-2}	3 mAh cm^{-2}	68	97.2	
Ni _x N layer on Nickel Foam (PNNF)	1 mA cm^{-2}	1 mAh cm^{-2}	300	97	
	2 mA cm^{-2}	1 mAh cm^{-2}	150	98.6	22
	5 mA cm^{-2}	1 mAh cm^{-2}	100	95.6	
3D Hollow Porous Cu Fibers (3D-HPCF)	0.5 mA cm^{-2}	1 mAh cm^{-2}	350	99.21	
	1 mA cm^{-2}	1 mAh cm^{-2}	300	97.8	23
	3 mA cm^{-2}	1 mAh cm^{-2}	150	97.26	
3D Cu-Cu _x O	1 mA cm^{-2}	1 mAh cm^{-2}	350	99.5	
	2 mA cm^{-2}	2 mAh cm^{-2}	215	99.3	This
	3 mA cm^{-2}	1 mAh cm^{-2}	170	99.2	Work
	5 mA cm^{-2}	1 mAh cm^{-2}	100	99.0	

Video S1. The Li-infusing experiment of Cu foam.

Video S2. The Li-infusing experiment of the Cu-Cu_xO host.

Video S3. In-situ optical microscopy observation of Li deposition on Cu foam under the current density of 0.5 mA (64 times speed playback).

Video S4. In-situ optical microscopy observation of Li deposition on the $Cu-Cu_xO$ host under the current density of 0.5 mA (64 times speed playback).

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