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Electronic Supplementary Information

Riveting the Atomically Distributed Lithiophilic Centers in the CNT Reinforced Interfacial Layer: An Ultra-thin, Light-weight Deposition Substrate toward Superior Li Utilization

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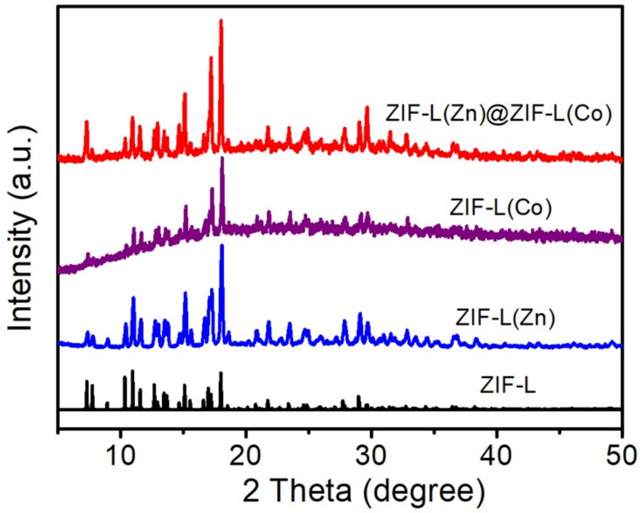

Fig. S1 XRD pattern of the ZIF-L(Zn)@ZIF-L(Co) precursor.

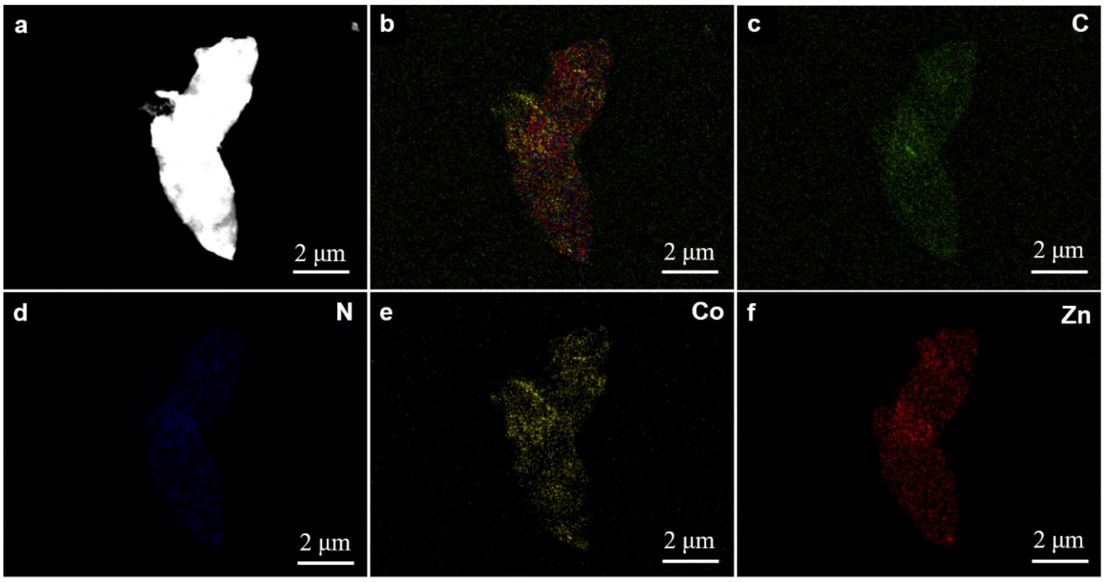

Fig. S2 SEM image and corresponding EDS elemental maps for the ZIF-L(Zn)@ZIF-L(Co) precursor.

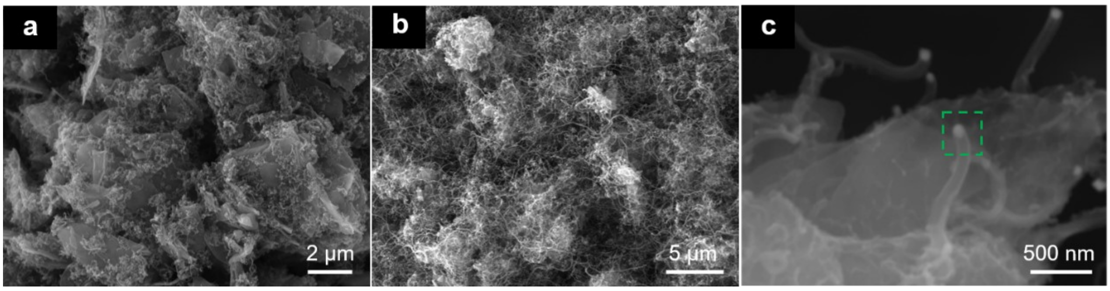

Fig. S3 SEM images of the Zn-NC-CNT composite at different magnifications. Co was represented in the dotted line in Figure S3c.

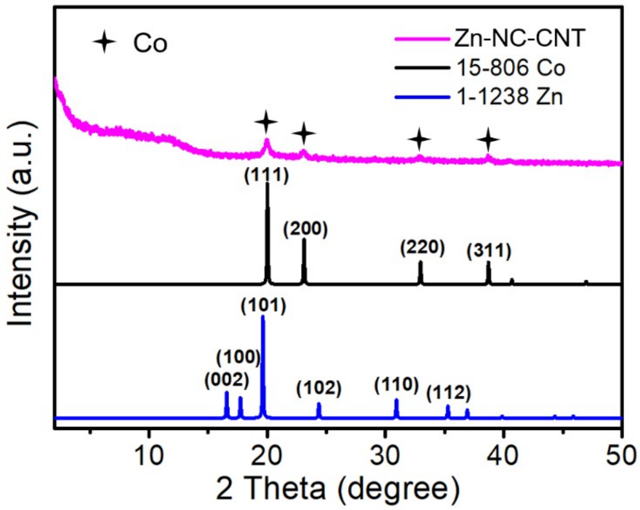

Fig. S4 XRD pattern of the Zn-NC-CNT composite.

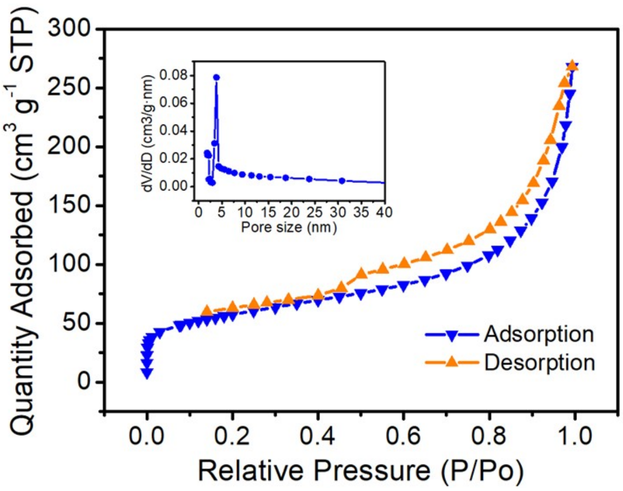

Fig. S5 Nitrogen adsorption-desorption isotherm and BJH pore size distribution (inset) of the Zn-NC-CNT composite.

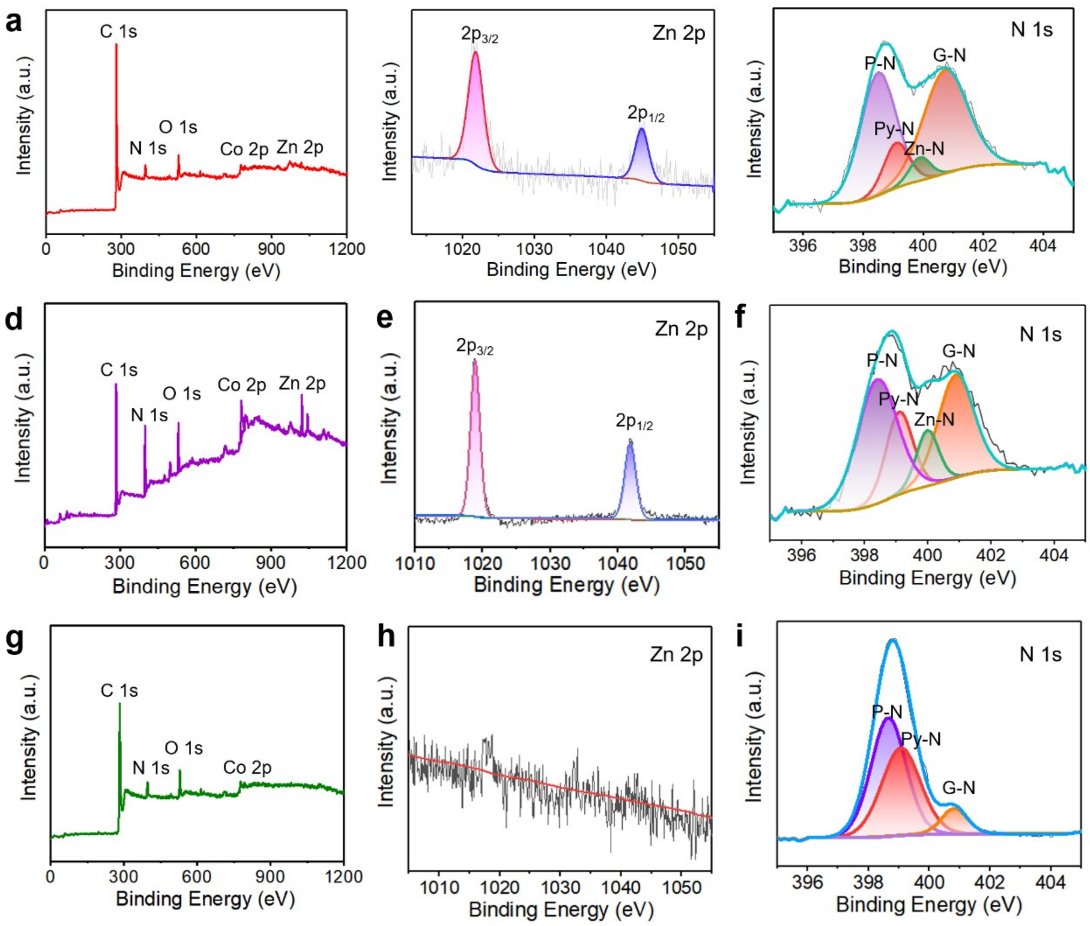

Fig. S6 (a,b,c) XPS spectra of the Zn-NC-CNT composite. (d,e,f) XPS spectra of the Zn-NC composite. (h,i,j) XPS spectra of the NC-CNT composite. P-N, Py-N, and G-N denote pyridinic N, pyrrolic N, and graphitic N, respectively.

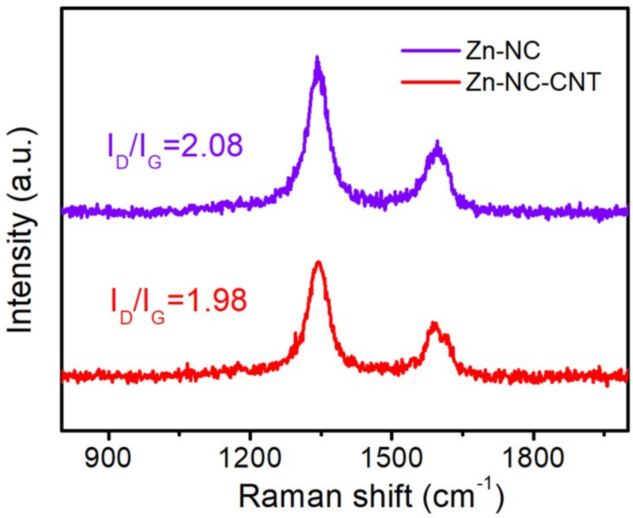

Fig. S7 Raman spectra of the Zn-NC-CNT and Zn-NC composites.

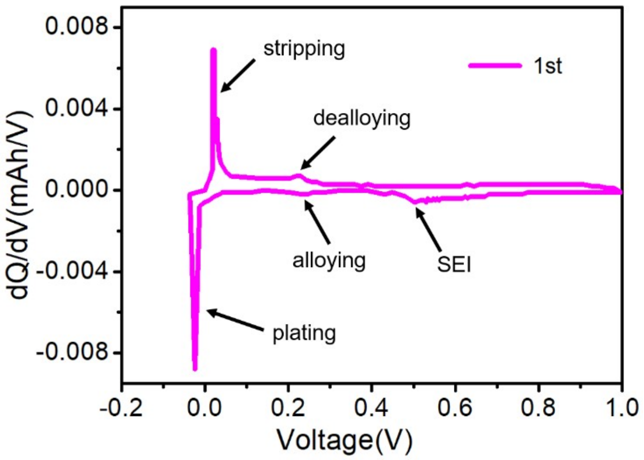

Fig. S8 The differential capacitance curve of the Zn-NC-CNT-Cu half cell for the first cycle.

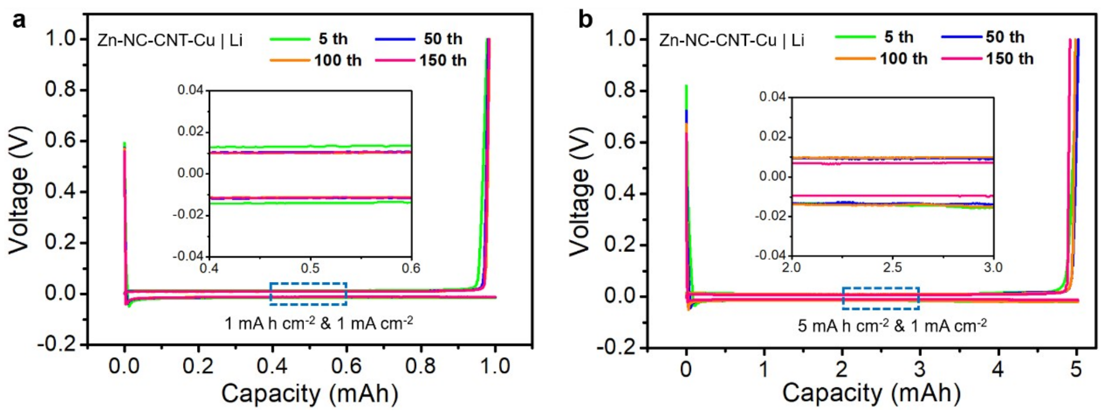

Fig. S9 Voltage profiles of the different cycles of Zn-NC-CNT-Cu electrode with (a) 1 mA h cm^{-2} capacity and (b) 5 mA h cm^{-2} capacity.

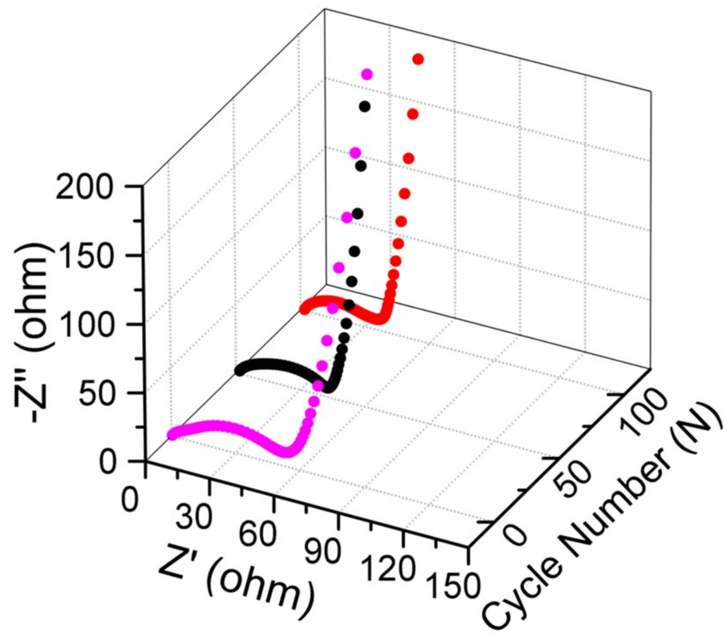

Fig. S10 Nyquist plots of the impedance spectra of Zn-NC-CNT-Cu symmetric cell before cycling, after 50 cycles and after 100 cycles.

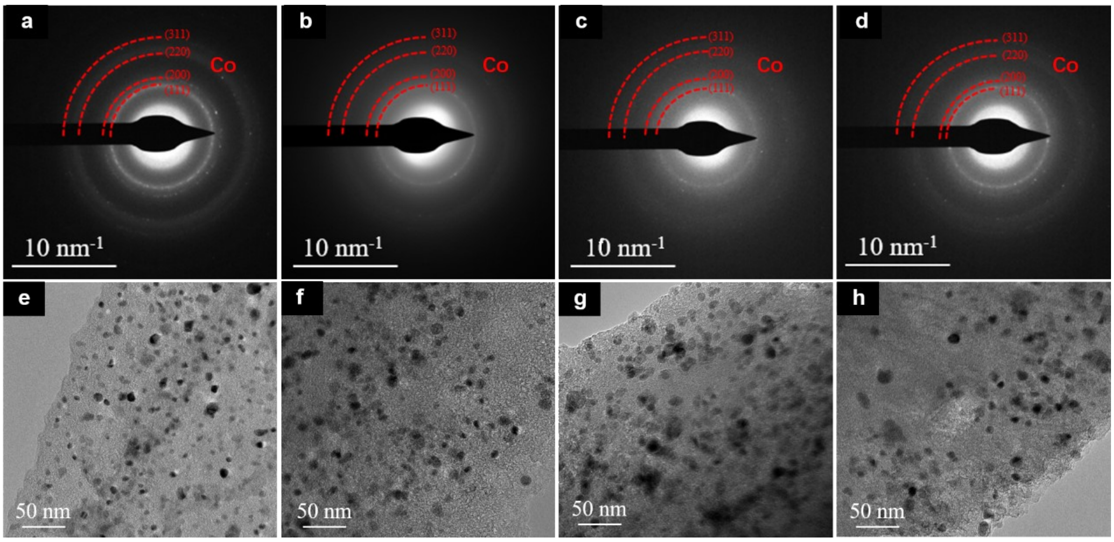

Fig. S11 (a) SAED and (e) HRTEM images of the Zn-NC-CNT-Cu electrode before cycle. (b) SAED and (f) HRTEM images of the Zn-NC-CNT-Cu electrode after discharge to 0 V. (c) SAED and (g) HRTEM images of the Zn-NC-CNT-Cu electrode after the lithium plating process. (d) SAED and (h) HRTEM images of the Zn-NC-CNT-Cu electrode after the lithium stripping process.

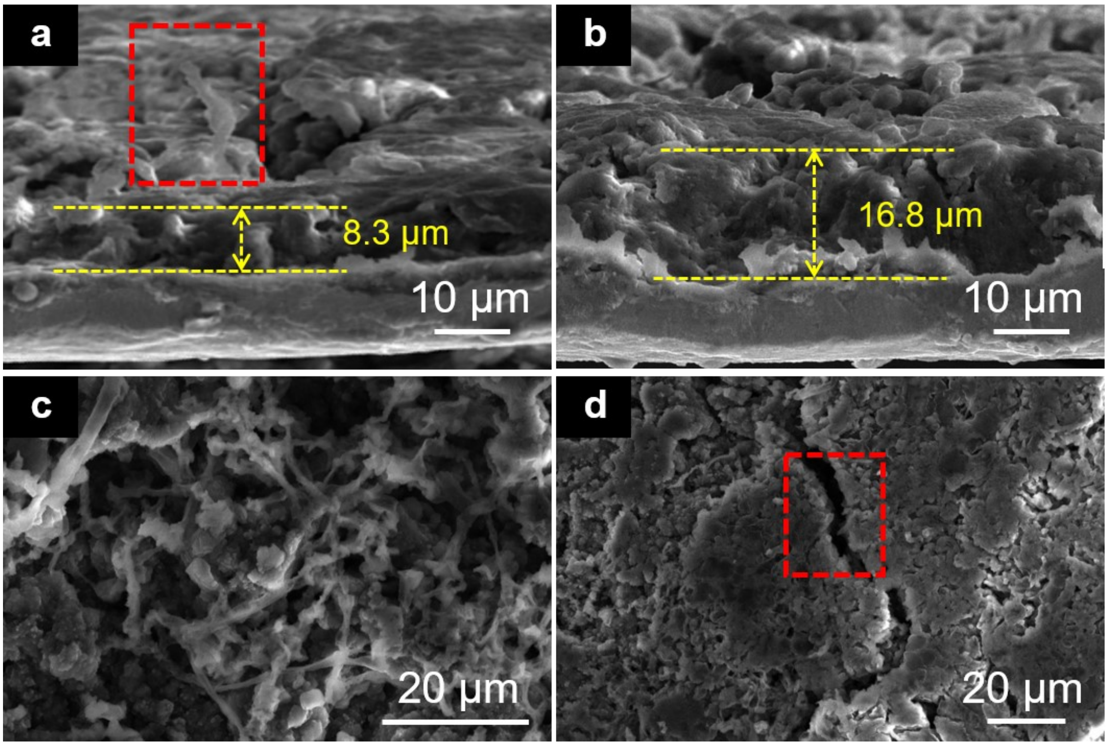

Fig. S12 (a) Cross section view and (c) top view SEM images of the NC-CNT-Cu electrode after 300 cycles of Li plating/stripping process with a 1 mA h cm^{-2} capacity limit. (b) Cross section view and (d) top view SEM images of the Zn-NC-Cu electrode after 300 cycles of Li plating/stripping process with 1 mA h cm^{-2} capacity.

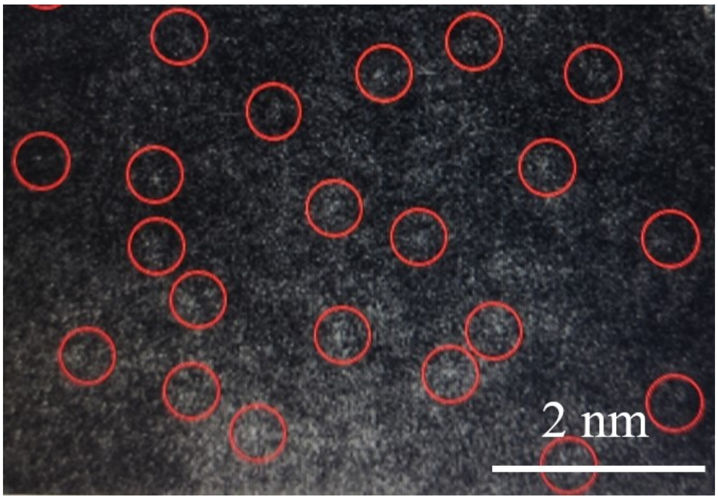

Fig. S13 Zoom-in HAADF-STEM image of the Zn-NC-CNT-Cu electrode after 300 cycles of Li plating/stripping process, in which the Zn species were highlighted with the red circles.

Supplementary Note:

The full cell prototype was constructed when pairing the Zn-NC-CNT-Cu (0 excess) with the NMC-811 cathode. The rated capacity was preset as 2 mA h. The mass loading of the Zn-NC-CNT-Cu (0 excess) composite on the copper foil (Φ12mm) was measured as 0.25 mg, the mass loading of the NMC-811 cathode on the aluminum foil (Φ12mm) was measured as 11.65 mg. A constant k (where $k = \frac{m_p + m_n}{m}$), defined as the mass fraction of active materials (*i.e.*, cathode and anode mass divided by total battery mass). The energy density and power density were calculated by below equations:

$$\begin{aligned} &\text{Energy } density\ (Wh\ kg^{-1}) \\ &= k \times \text{specific capacity (mA h g}^{-1}) \times \text{average voltage (V)} \\ &= k \times \frac{\text{capacity of the full cell } (mA\ h)\ \times \text{average voltage (V)}}{\text{weight of (anode + cathode)} (mg)} \end{aligned} \quad (1)$$

$$\begin{aligned} &\text{Power density (W kg}^{-1}) \\ &= \text{Energy density } (Wh\ kg^{-1}) \times \text{ C rate (h}^{-1}) \end{aligned} \quad (2)$$

When the energy density of the Zn-NC-CNT-Cu (0 excess)||NMC-811 full cell was calculated based on the loading mass of active materials ($m_p + m_n = m$), the constant k was 1 (ideal condition), as shown below:

When the current density was increased to 0.5 C, the energy density and power density were 638.8 Wh $kg^{-1}$and 319.39 W kg^{-1}.

Specific capacity $= \frac{2 \text{ mA h}}{0.25\ mg + 11.65 \text{ mg}} =$ 168.1 mA h g^{-1}.

Energy density = (168.1 mA h g^{-1}) × (3.8 V) = 638.8 Wh kg^{-1}.

Power density = (638.8 Wh kg^{-1}) × (0.5 h^{-1}) = 319.4 W kg^{-1}.

When the current density was increased to 1 C, the energy density and power density were 606.9 Wh kg^{-1}and 606.9 W kg^{-1}.

Specific capacity $= \frac{1.90 \text{ mA h}}{0.25\ mg + 11.65 \text{ mg}} =$ 159.7 mA h g^{-1}.

Energy density = (159.7 mA h g^{-1}) × (3.8 V) = 606.9 Wh kg^{-1}.

Power density = (606.9 Wh kg^{-1}) × (1 h^{-1}) = 606.9 W kg^{-1}.

When the current density was increased to 2 C, the energy density and power density were 581.4 Wh kg^{-1}and 1162.8 W kg^{-1}.

Specific capacity $= \frac{1.82 \text{ mA h}}{0.25\ mg + 11.65 \text{ mg}} =$ 153.0 mA h g^{-1}.

Energy density = (153.0 mA h g^{-1}) × (3.8 V) = 581.4 Wh kg^{-1}.

Power density = (581.4 Wh kg^{-1}) × (2 h^{-1}) = 1162.8 W kg^{-1}.

When the current density was increased to 3 C, the energy density and power density were 555.6 Wh kg^{-1}and 1666.8 W kg^{-1}.

Specific capacity $= \frac{1.74 \text{ mA h}}{0.25\ mg + 11.65 \text{ mg}} =$ 146.2 mA h g^{-1}.

Energy density = (146.2 mA h g^{-1}) × (3.8 V) = 555.6 Wh kg^{-1}.

Power density = (555.6 Wh kg^{-1}) × (3 h^{-1}) = 1666.8 W kg^{-1}.

When the current density was increased to 4 C, the energy density and power density were 530.1 Wh kg^{-1}and 319.39 W kg^{-1}.

Specific capacity $= \frac{1.66 \text{ mA h}}{0.25\ mg + 11.65 \text{ mg}} =$ 139.5 mA h g^{-1}.

Energy density = (139.5 mA h g^{-1}) × (3.8 V) = 530.1 Wh kg^{-1}.

Power density = (530.1 Wh kg^{-1}) × (4 h^{-1}) = 2120.4 W kg^{-1}.

Table S1. The calculation results of the specific capacity, energy density and power density of the Zn-NC-CNT-Cu (0 excess)||NMC-811 full cell at different C rates (based on the active materials).

	Zn-NC-CNT-Cu (0 excess)||NMC-811		
C rate (h^{-1})	Specific capacity (mA h g^{-1})	Energy density (Wh kg^{-1})	Power density (W kg^{-1})
0.5	168.1	638.8	319.4
1	159.7	606.9	606.9
2	153.0	581.4	1162.8
3	146.2	555.6	1666.8
4	139.5	530.1	2120.4

We also calculated the specific capacity, energy density and power density of the Cu||NMC-811 and Li||NMC-811 full cells at different C rates based on the electrode active material, as summarized in Table S2 and S3.

Table S2. The calculation results of the specific capacity, energy density and power density of the Cu||NMC-811 full cell at different C rates (based on the active materials).

	Cu||NMC-811		
C rate (h^{-1})	Specific capacity (mA h g^{-1})	Energy density (Wh kg^{-1})	Power density (W kg^{-1})
0.5	170.9	649.4	324.7
1	116.9	444.2	444.2
2	75.4	286.5	573.0
3	47.1	179.0	537.0
4	28.1	106.8	427.2

Table S3. The calculation results of the specific capacity, energy density and power density of the Li||NMC-811 full cell at different C rates (based on the active materials).

	Li||NMC-811		
C rate (h^{-1})	Specific capacity (mA h g^{-1})	Energy density (Wh kg^{-1})	Power density (W kg^{-1})
0.5	140.6	533.3	266.7
1	136.8	520.0	520.0
2	133.3	506.6	1013.3
3	129.8	493.3	1480.0
4	126.3	480.0	1920.1

We can use these empirical values (*i.e.*, k = 0.45-0.55) to estimate the energy density of the Zn-NC-CNT-Cu (0 excess)||NMC-811 full cell. And we set the parameters k equal to 0.55, the related results are summarized in Table S4.

Table S4. The calculation results of the specific capacity, energy density and power density of the Zn-NC-CNT-Cu (0 excess)||NMC-811 full cell at different C rates (based on the whole mass of the cell for practical battery applications, while k= ~ 0.55).

	Zn-NC-CNT-Cu (0 excess)||NMC-811		
C rate (h^{-1})	Specific capacity (mA h g^{-1})	Energy density (Wh kg^{-1})	Power density (W kg^{-1})
0.5	168.1	351.34	175.67
1	159.7	333.795	333.795
2	153.0	319.77	639.54
3	146.2	305.58	916.74
4	139.5	291.555	1166.22