Supplementary Data

Stabilization of Lithium Metal Anodes by Conductive Metal-Organic Framework Architectures

Jing Zhao,[ab] Hongye Yuan,[a] Guiling Wang,*[b] Xiao Feng Lim,[a] Hualin Ye,[a] Vanessa Wee,[a] Yongzheng Fang,[b] Jim Yang Lee,*[a] Dan Zhao,*[a]

[a] *Department of Chemical and Biomolecular Engineering, National University of Singapore, Singapore 117585, Singapore*

[b] *Key Laboratory of Superlight Materials and Surface Technology of Ministry of Education, Department of Materials Science and Engineering, Harbin Engineering University, Harbin, Heilongjiang, 150001, P. R. China*

Corresponding Author

[a,*] *Dan Zhao* E-mail: chezhao@nus.edu.sg, *Jim Yang Lee* E-mail: cheleejy@nus.edu.sg.

[b,*] *Guiling Wang* E-mail: wangguiling@hrbeu.edu.cn.

Table of Contents

1. **Supplementary Table** S3
2. **Supplementary Figures** S4

1. Supplementary Table

Table S1. The values of adsorption energy and diffusion barrier of Li ions on different surfaces calculated by the DFT method.

Sample	Adsorption energy (eV Å^{-2})	Diffusion barrier (eV Å^{-2})
Cu (111)	-2.37	1.76
$Ni_3(HITP)_2$@Cu-foil	-3.18	0.71

2. Supplementary Figures

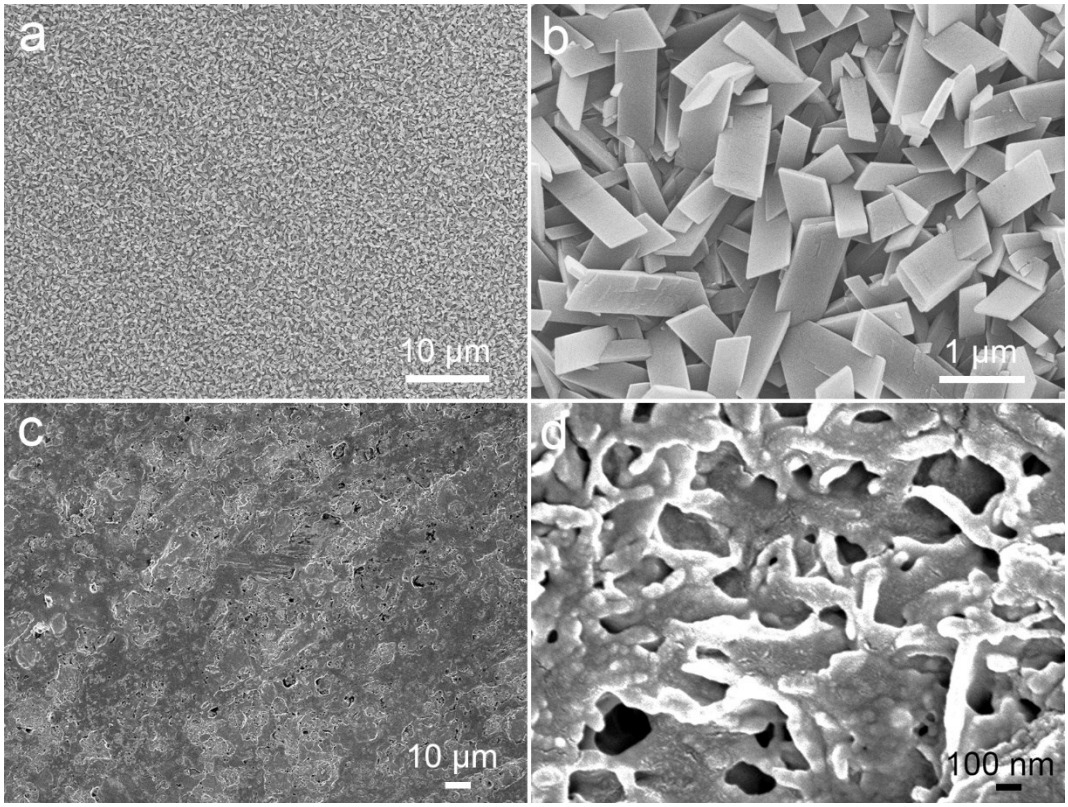

Fig. S1. SEM images of Ni-NDC@Cu-foil current collector: (a) low magnification, (b) high magnification. SEM images of $Ni_3(HITP)_2$@Cu-foil after cycling: (c) low magnification, (d) high magnification.

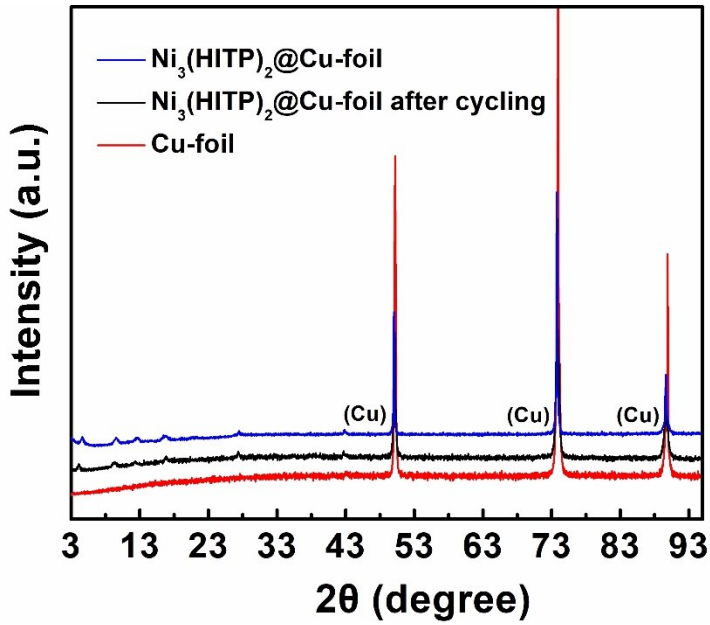

Fig. S2. Full XRD patterns of different current collectors.

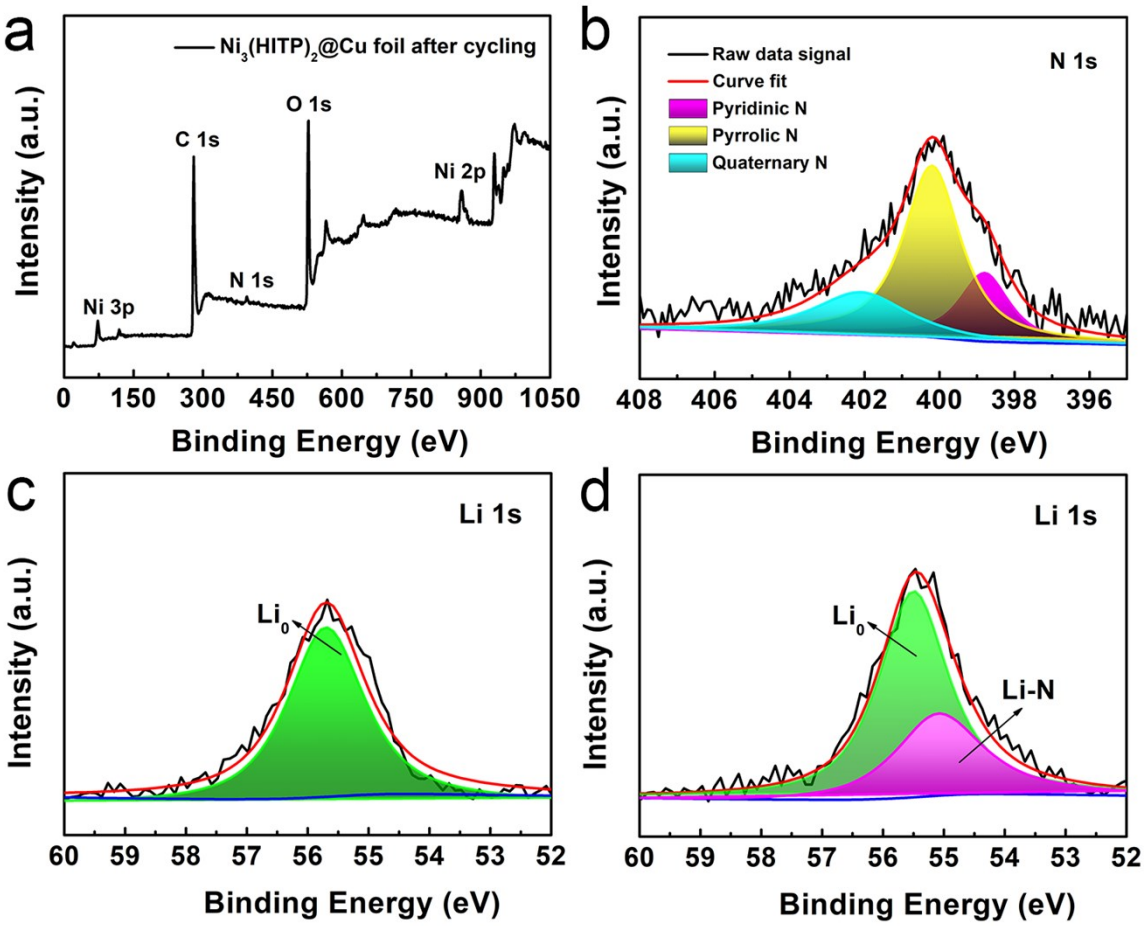

Fig. S3. (a) Wide-scan XPS spectra and (b) high-resolution N 1s XPS spectra of the $Ni_3(HITP)_2$@Cu-foil after cycling. Li 1s XPS spectra of (c) initial $Ni_3(HITP)_2$@Cu-foil and (d) Li/$Ni_3(HITP)_2$@Cu-foil.

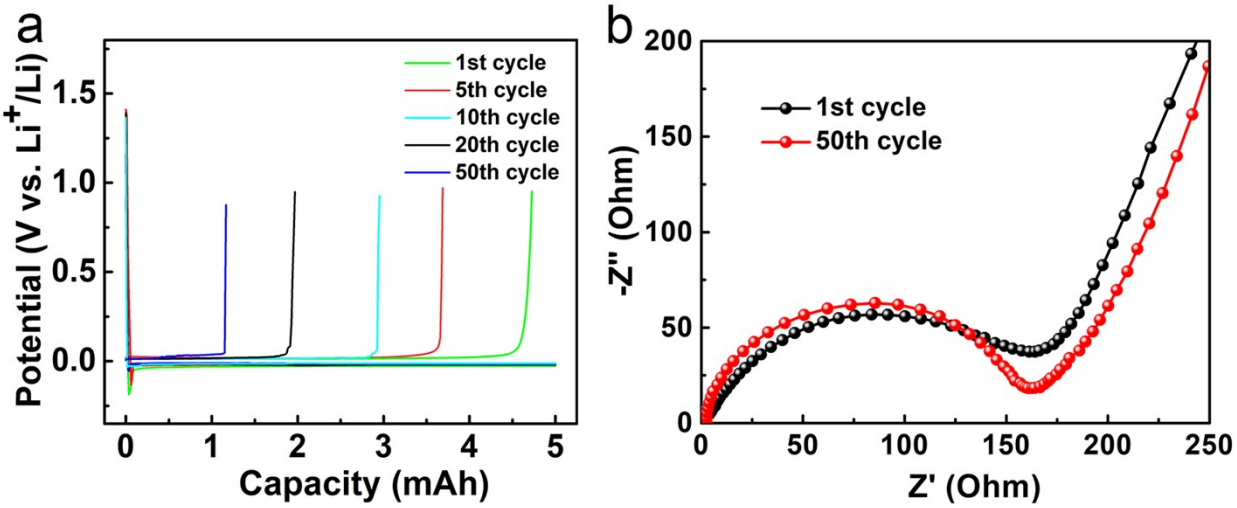

Fig. S4. (a) Voltage profiles of Li plating/stripping on/from planar Cu-foil at a current density of 5 mA cm^{-2} with an areal capacity of 5 mAh cm^{-2} after different cycles. (b) EIS plots of $Ni_3(HITP)_2$@Cu-foil at different cycles.

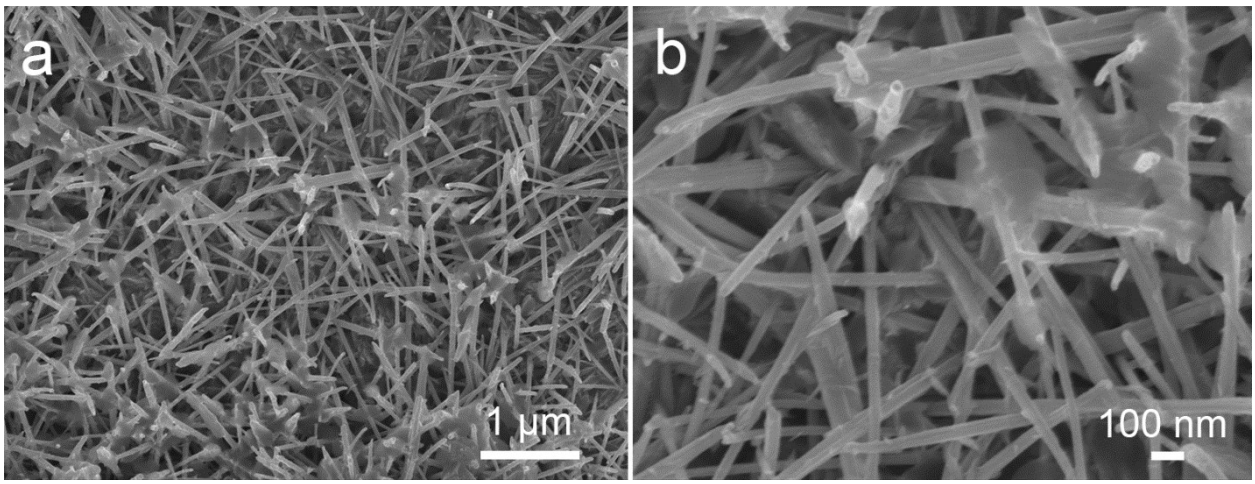

Fig. S5. SEM images of lithium metal deposited on the planar Cu-foil current collector under (a) low magnification and (b) high magnification.

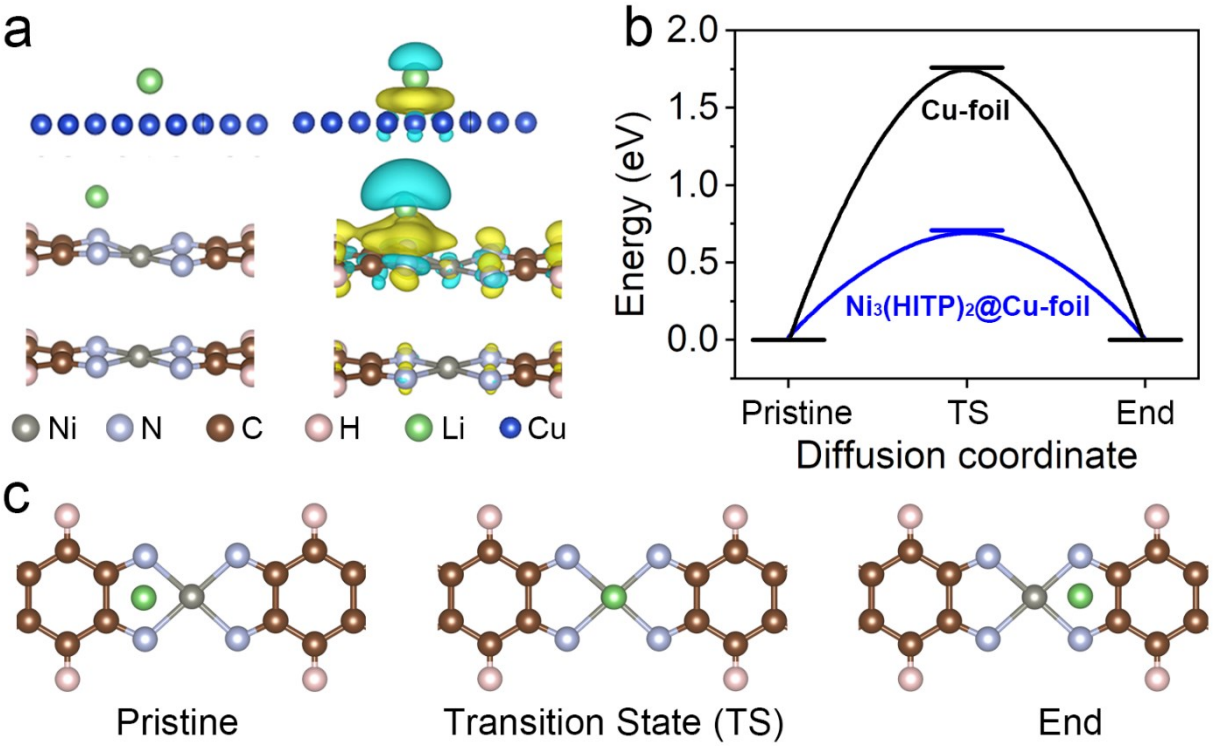

Fig. S6. a) Geometric configuration and corresponding charge density differences of a Li atom on the Cu-foil and $Ni_3(HITP)_2$@Cu-foil (the yellow and cyan regions represent positive and negative densities respectively. The isovalue for the charge density was taken as ± 0.002 e $Å^{-3}$). b) A comparison of the diffusion barriers on Cu (111) and $Ni_3(HITP)_2$. c) The diffusion pathway of Li atoms through the $Ni_3(HITP)_2$ layers.

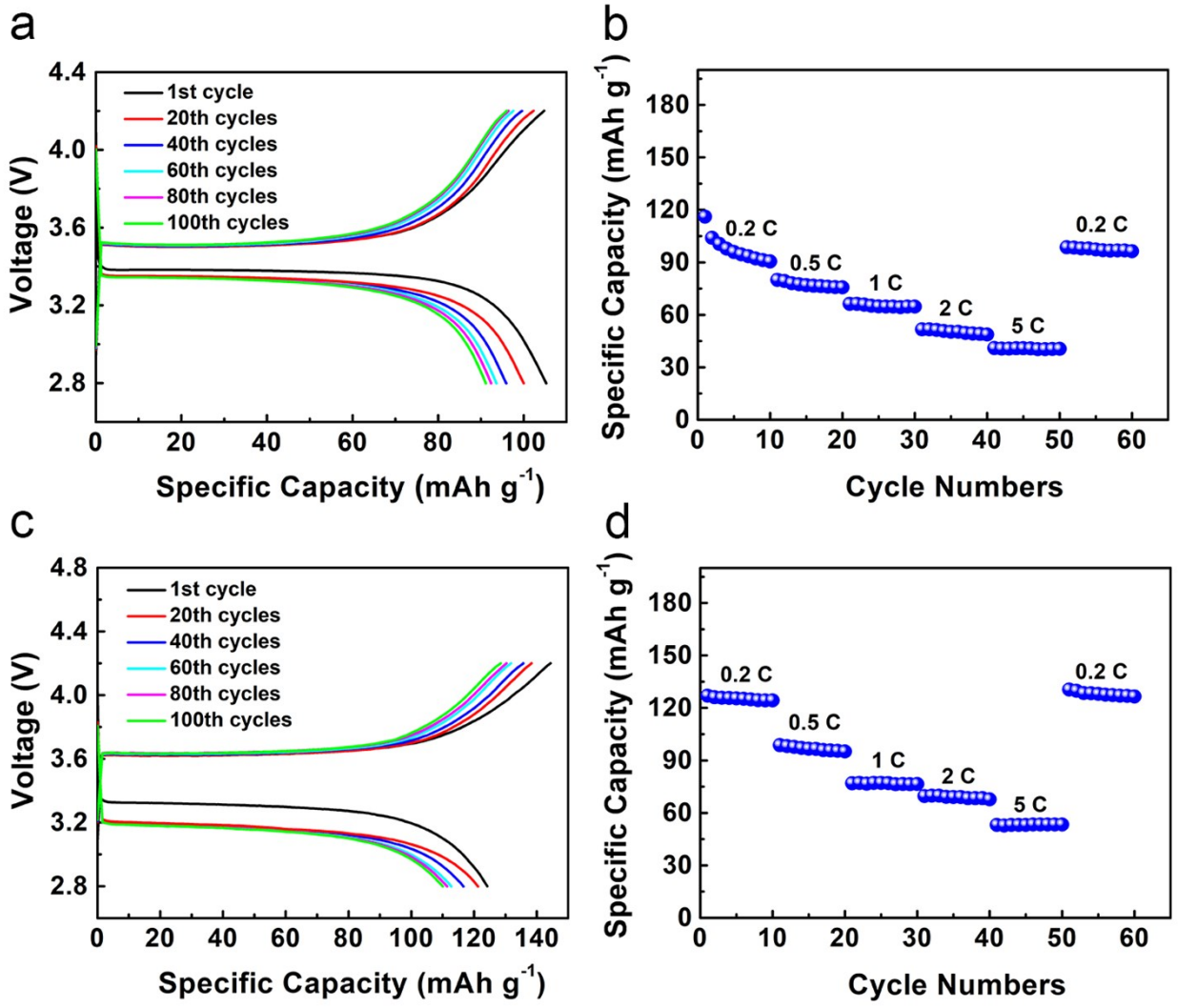

Fig. S7. (a) Voltage profiles and (b) rate capability of the Li/$Ni_3(HITP)_2$@Cu-foil|LFP cells with LFP capacity of 1.5 mAh cm^{-2}. (c) Voltage profiles and (d) rate capability of the Li/$Ni_3(HITP)_2$@Cu-foil|LFP cells with LFP capacity of 2.4 mAh cm^{-2}.