

## Supporting Information

# Coupling of Triporosity and Strong Au-Li Interaction to Enable Dendrite-Free Lithium Plating/Stripping for Long-Life Lithium Metal Anodes

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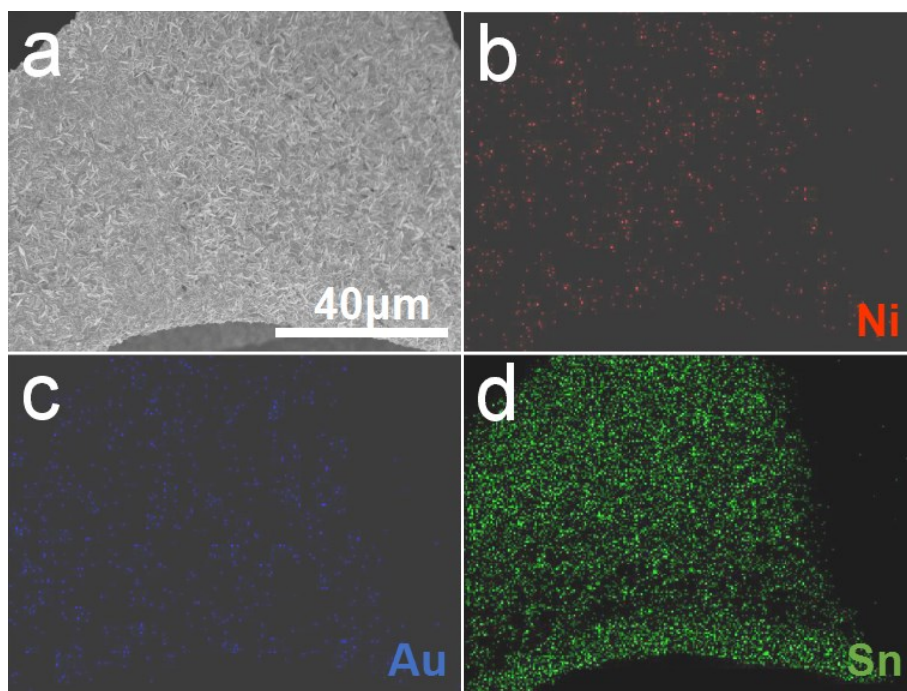
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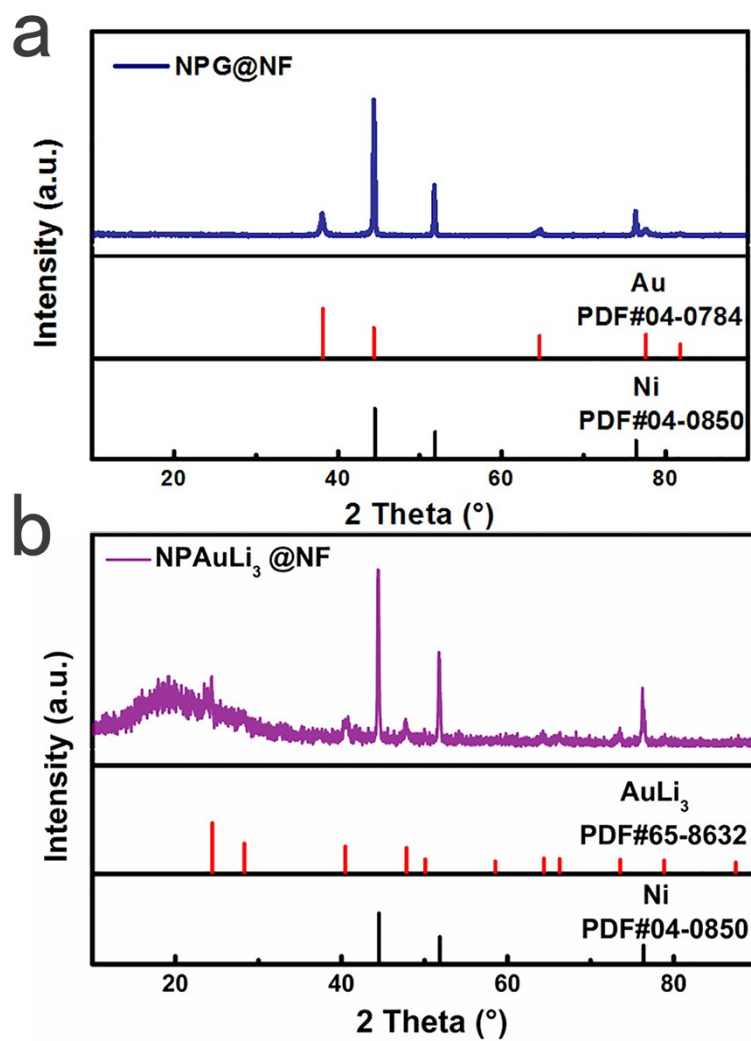
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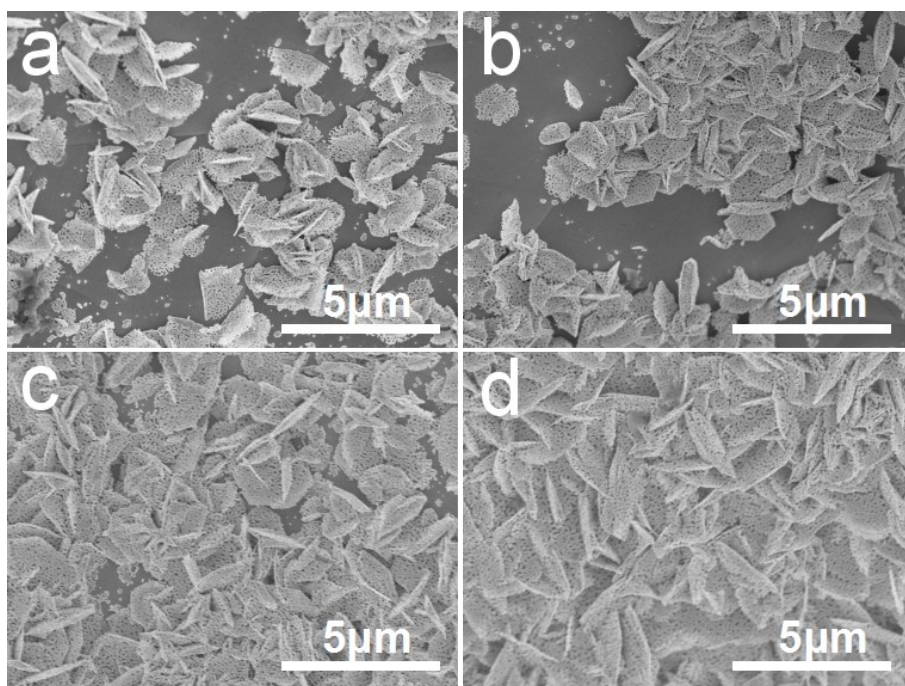
<sup>1</sup> Y.H. Liang and Y.M. Chen contributed equally to this work.



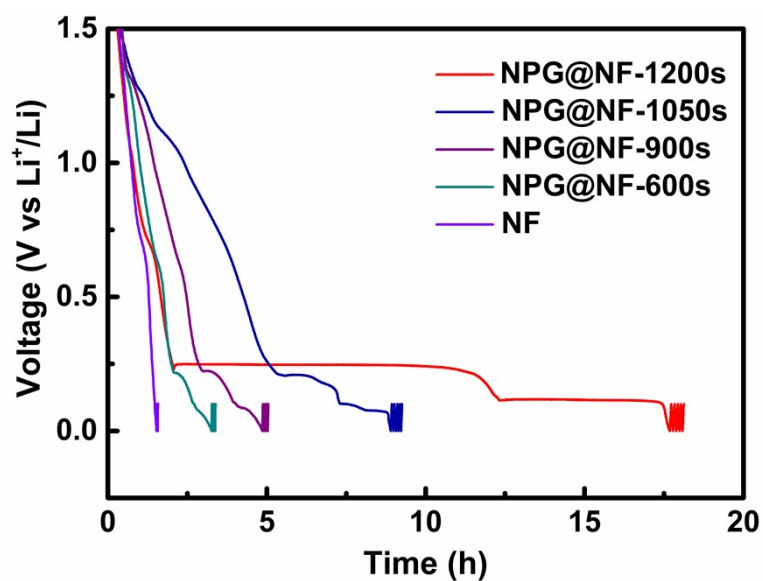
**Figure S1.** SEM image of AuSn@NF electrode (a) and the corresponding EDX elemental mapping images of Ni (b), Au (c) and Sn (d).



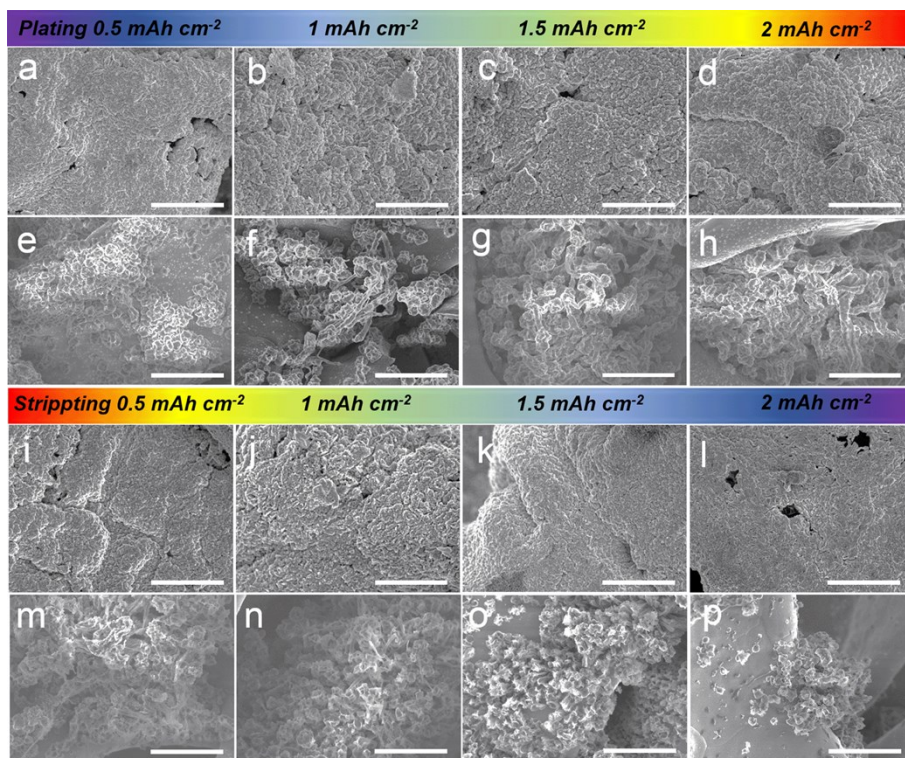
**Figure S2.** XRD patterns of NPG@NF (a) and NPAuLi<sub>3</sub>@NF (b) electrodes.



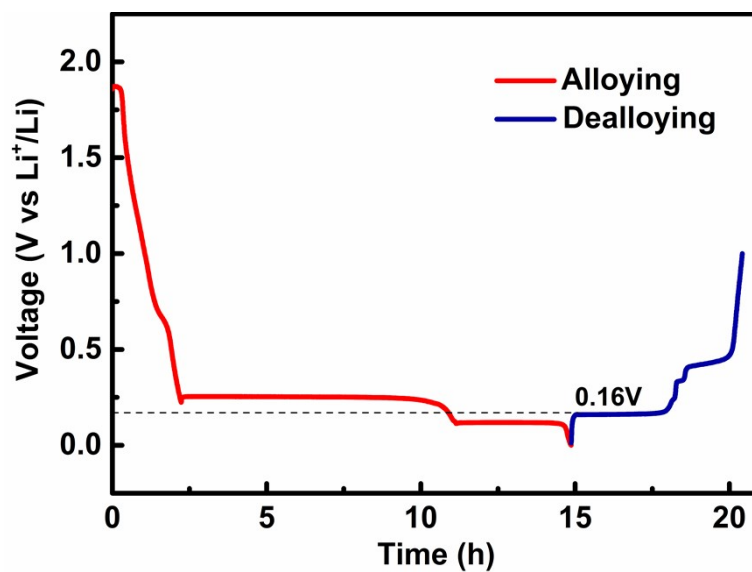
**Figure S3.** SEM images of NPG@NF obtained by chemically dealloying AuSn@NF electrodes electrodeposited at a current density of  $5 \text{ mA cm}^{-2}$  for 600 (a), 900 (b), 1050 (c) and 1200 (d) s, respectively.



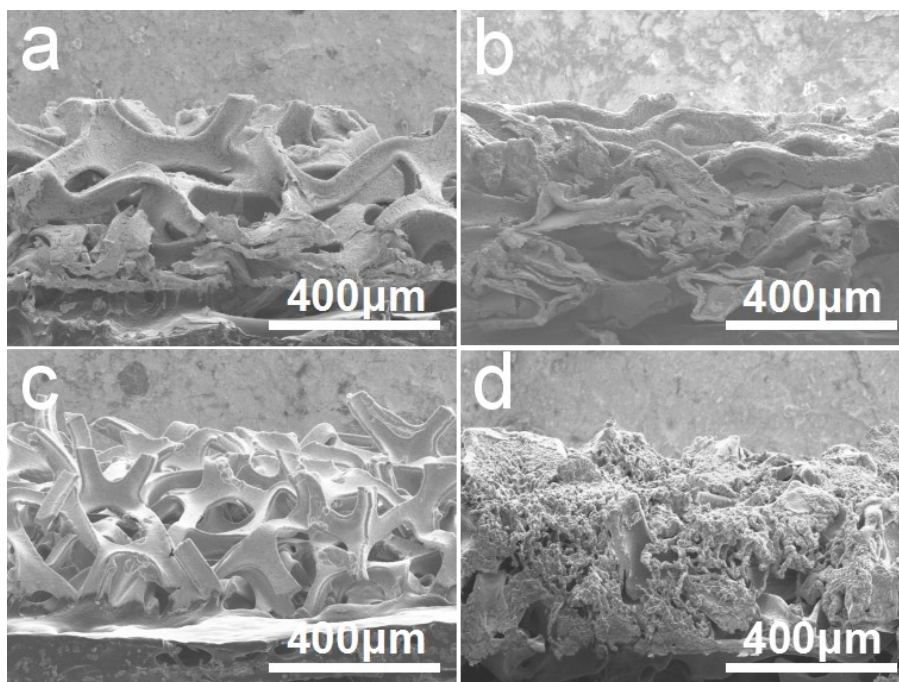
**Figure S4.** Voltage profiles of a galvanostatic discharge process to 0 V (vs.  $\text{Li}^+/\text{Li}$ ) followed by a cycling between 0 and 0.1 V for 5 cycles at a current density of  $50 \mu\text{A cm}^{-2}$  for NF and NPG@NF electrodes. NPG@NF electrodes were obtained by chemically dealloying AuSn@NF electrodes electrodeposited at a current density of  $5 \text{ mA cm}^{-2}$  for 600 (NPG@NF-600s), 900 (NPG@NF-900s), 1050 (NPG@NF-1050s) and 1200 (NPG@NF-1200s) s, respectively.



**Figure S5.** SEM images of NPAuLi<sub>3</sub>@NF (a-d, i-l) and pristine NF (e-h, m-p) electrodes after plating 0.5 (a, e), 1 (b, f), 1.5 (c, g) and 2 (d, h) mAh cm<sup>-2</sup> Li, and after stripping 0.5 (i, m), 1 (j, n), 1.5 (k, o) and 2 (l, p) mAh cm<sup>-2</sup> Li, respectively, at a current density of 0.5 mA cm<sup>-2</sup>. Scale bars: 20 μm.

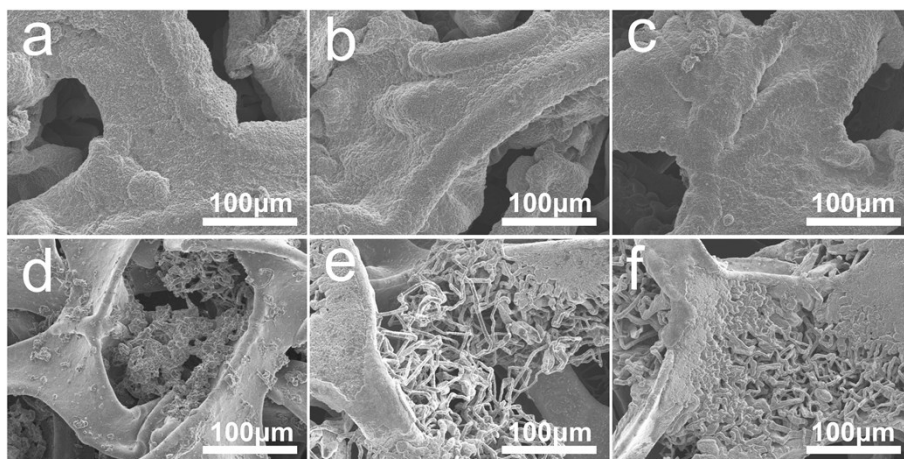


**Figure S6.** A representative voltage profile of the first discharge/charge cycle for a NPG@NF electrode at a current density of  $50 \mu\text{A cm}^{-2}$ .

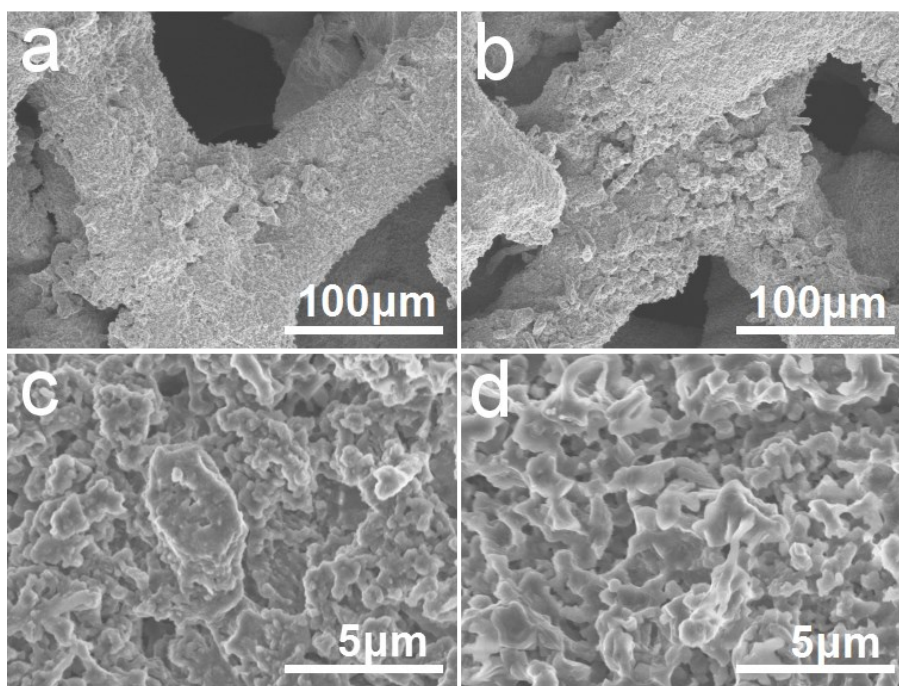


**Figure S7.** Cross-sectional SEM images of NPAuLi<sub>3</sub>@NF (a, b) and NF (c, d) electrodes before (a, c) and after 50 (b, d) plating/stripping cycles at a current density of 1 mA cm<sup>-2</sup> with a capacity of 2 mAh cm<sup>-2</sup>.

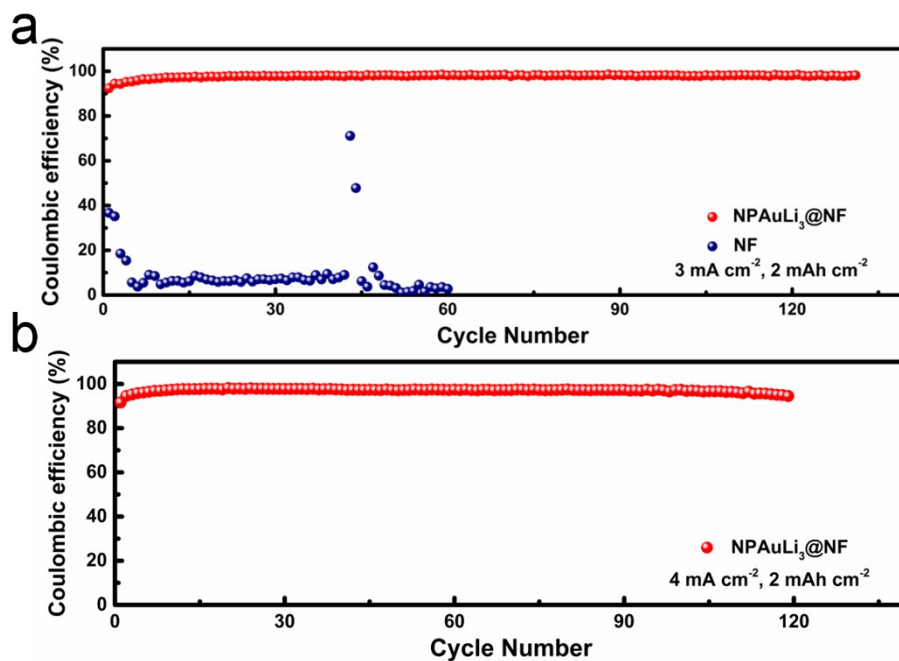




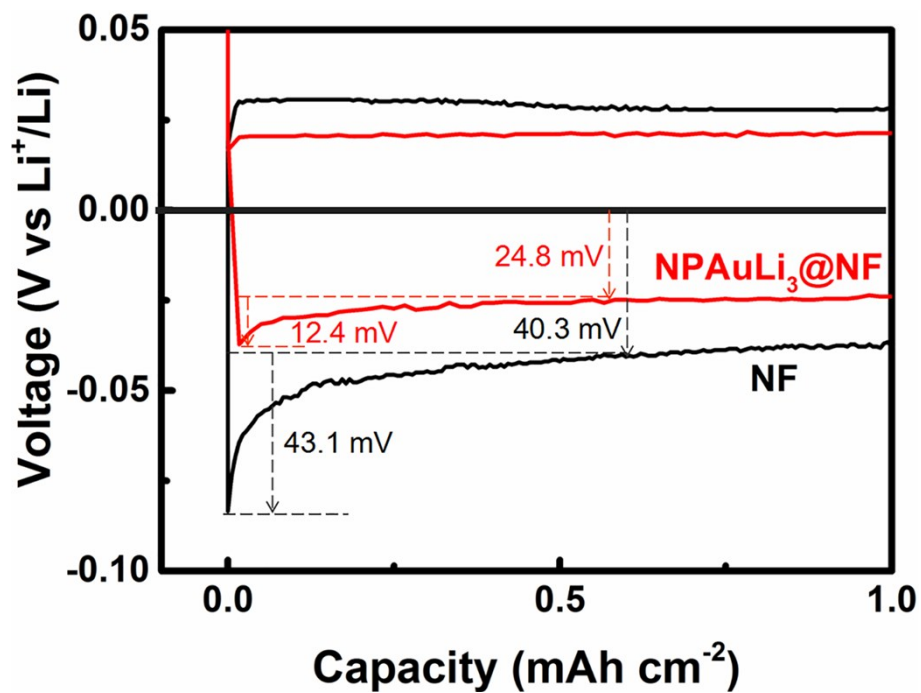
**Figure S8.** SEM images of the of the NPAuLi<sub>3</sub>@NF (a-c) and bare NF (d-f) electrodes after 10 (a, d), 50 (b, e) and 100 (c, f) plating/stripping cycles, respectively, at a current density of 2 mA cm<sup>-2</sup> with a capacity of 2 mAh cm<sup>-2</sup>.



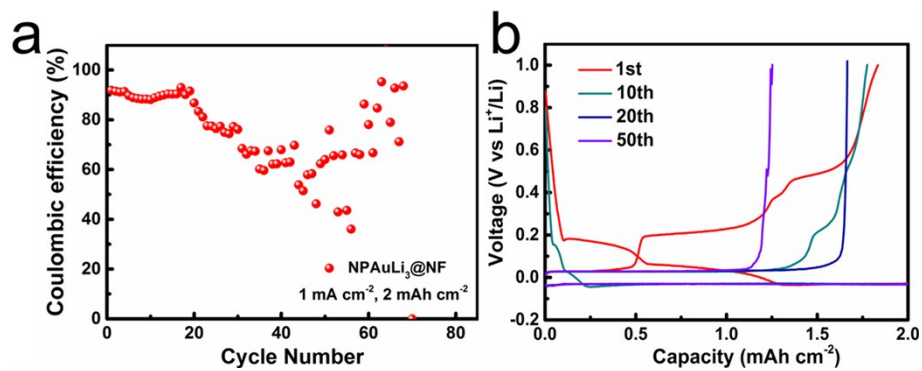
**Figure S9.** SEM images of NPAuLi<sub>3</sub>@NF electrode after 10 (a, c) and 50 (b, d) plating/stripping cycles under an upper cutoff voltage of 1.0 V at a current density of 1 mA cm<sup>-2</sup> with a capacity of 2 mAh cm<sup>-2</sup>.



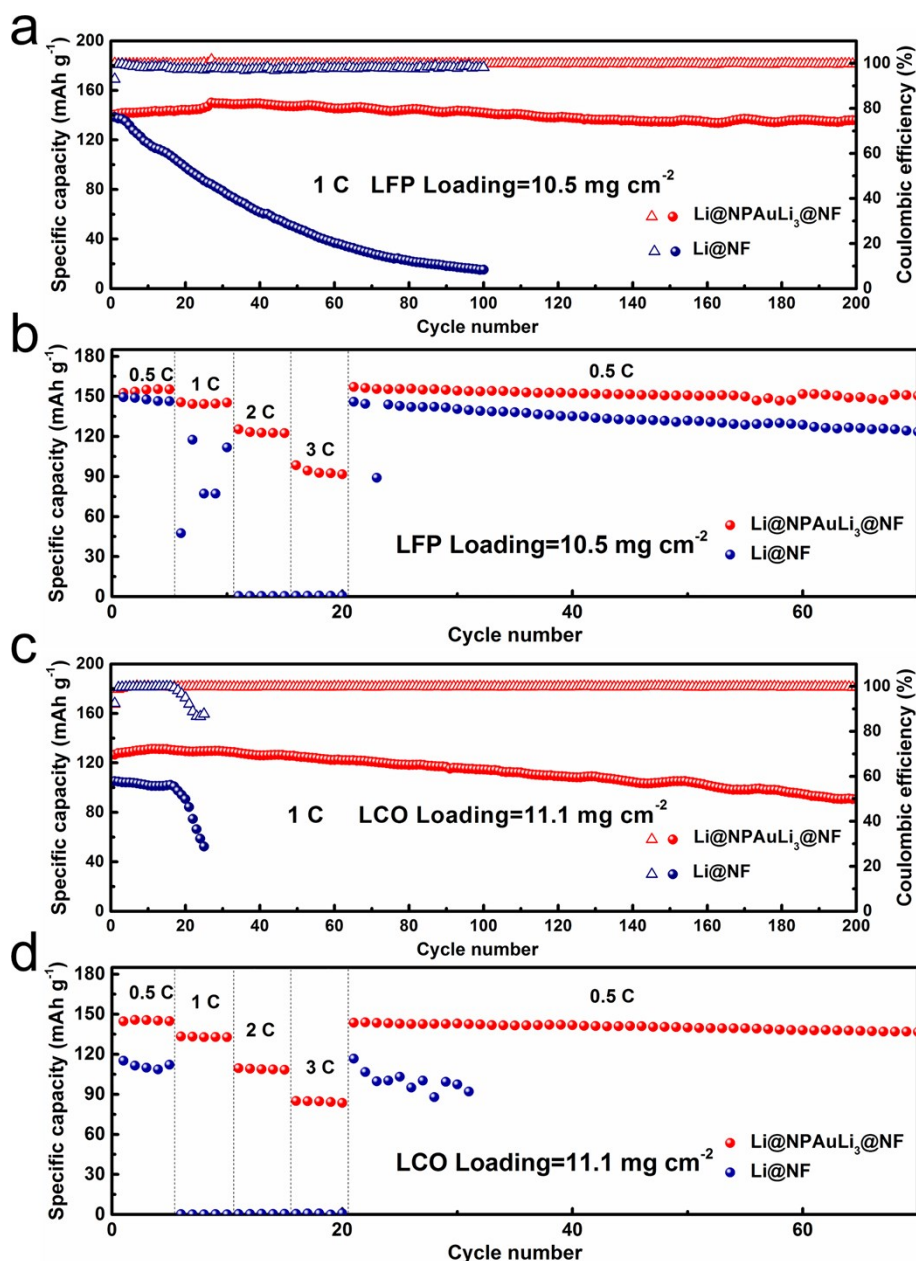
**Figure S10.** Coulombic efficiencies (CEs) for Li plating/stripping cycling on NPAuLi<sub>3</sub>@NF (a, b) and NF (a) electrodes at current densities of 3 (a) and 4 (b) mA cm<sup>-2</sup> with a capacity of 2 mAh cm<sup>-2</sup>.



**Figure S11.** Voltage profiles of galvanostatic Li deposition on bare NF and NPAuLi<sub>3</sub>@NF current collectors, respectively, at a current density of 1 mA cm<sup>-2</sup>.



**Figure S12.** CE (a) and voltage profiles (b) for Li plating/stripping cycles on the NPAuLi<sub>3</sub>@NF electrode under an upper cutoff voltage of 1.0 V at a current density of 1 mA cm<sup>-2</sup> with a capacity of 2 mAh cm<sup>-2</sup>.



**Figure S13.** Cycling performance of LMAs with bare NF and NPAuLi<sub>3</sub>@NF current collectors in full cells with a LiFePO<sub>4</sub> (LFP) (a) cathode with an areal mass loading of 10.5 mg cm<sup>-2</sup>, and a LiCoO<sub>2</sub> (LCO) (c) cathode with an areal mass loading of 11.1 mg cm<sup>-2</sup> at 1C. Rate capability of LMAs with bare NF and NPAuLi<sub>3</sub>@NF current collectors in full cells with a LFP (b) and LCO (d) cathode at different rates from 0.5 to 3C.

**Table S1.** Calculation results of Li atom adsorption on AuLi<sub>3</sub> and NF slabs

<b>Surface</b>	<b>Adsorption energies /eV</b>	<b>Adsorption site</b>
<b>Li-terminated AuLi<sub>3</sub> slab</b>	-1.81	fcc
<b>Au-terminated AuLi<sub>3</sub> slab</b>	-2.29	fcc
<b>NF slab</b>	-1.23	fcc

**Table S2.** Coordination and bond length of the adsorbed Li atom on different slabs

<b>Surface</b>	<b>Coordination number</b>	<b>Bond length /nm</b>
<b>Li-terminated</b>	3 Li atoms,	0.297 (Li-Li)
<b>AuLi<sub>3</sub> slab</b>	1 Au atom	0.265 (Li-Au)
<b>Au-terminated</b>		
<b>AuLi<sub>3</sub> slab</b>	3 Au atoms	0.2711 (Li-Au)
<b>NF slab</b>	3 Ni atoms	0.2556 (Li-Ni)



**Table S3.** Comparison of electrochemical performances of different LMAs in Li|Li symmetric cells

Anodes	Current density (mA cm <sup>-2</sup> )	Capacity (mAh cm <sup>-2</sup> )	Cycle time (h)	Ref.
Cu-CuO-Ni	0.5	0.5	580	1
3D Cu foil	0.2	1	1000	2
Cu@NPCN	0.5	1	1600	3
	1		1150	
NGCF@Li	2	1	1200	4
	3		600	
TiC/C/Li	1	1	400	5
	3		130	
Crumpled Graphene Balls@Li	0.5	1	750	6
AgNP/CNFs	0.5	1	500	7
GT scaffold	1	2	900	8
Li@CuCF	5	1	80	9
LNCO/Ni	1	1	1000	10
CNF-TiN	1	1	600	11
Li@Co <sub>3</sub> N/NF	1	1	1100	12
Li/CuNW-P	1	1	1000	13
NPAuLi <sub>3</sub> @NF	1	2	1990	This work
	2		1420	

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