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Supplementary Information

3D Conductive Scaffold with Lithiophilic Modification for Stable Lithium Metal Battery

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Figures

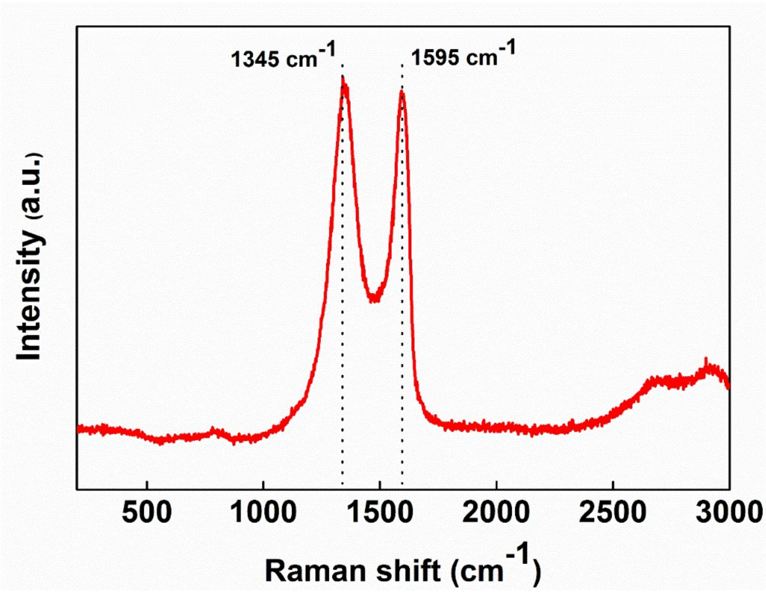

Fig. S1† Raman spectrum of NGNF electrode.

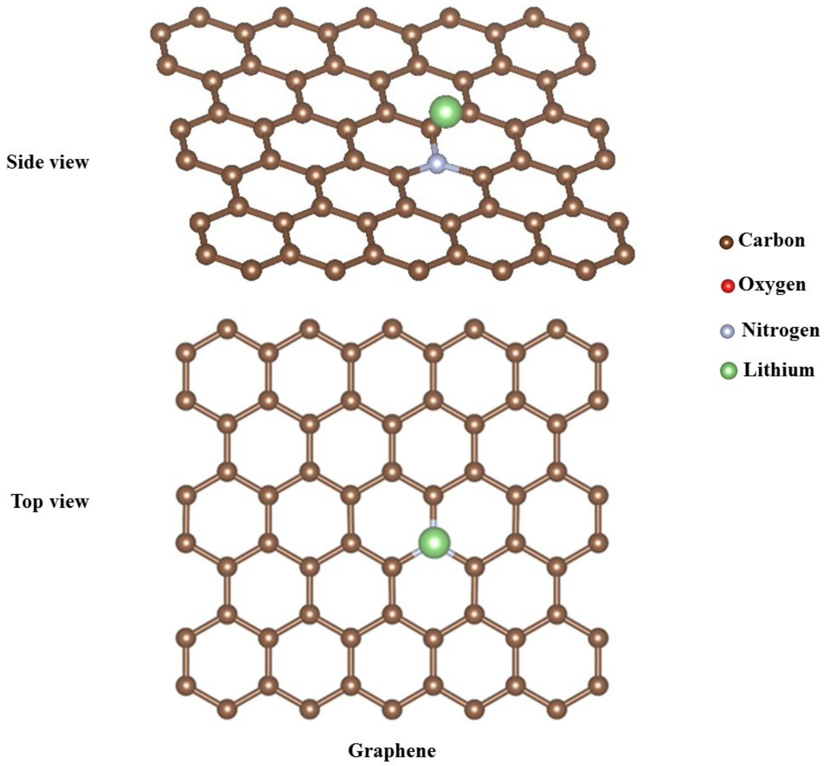

Fig. S2† Optimized structure of Li on ideal graphene.

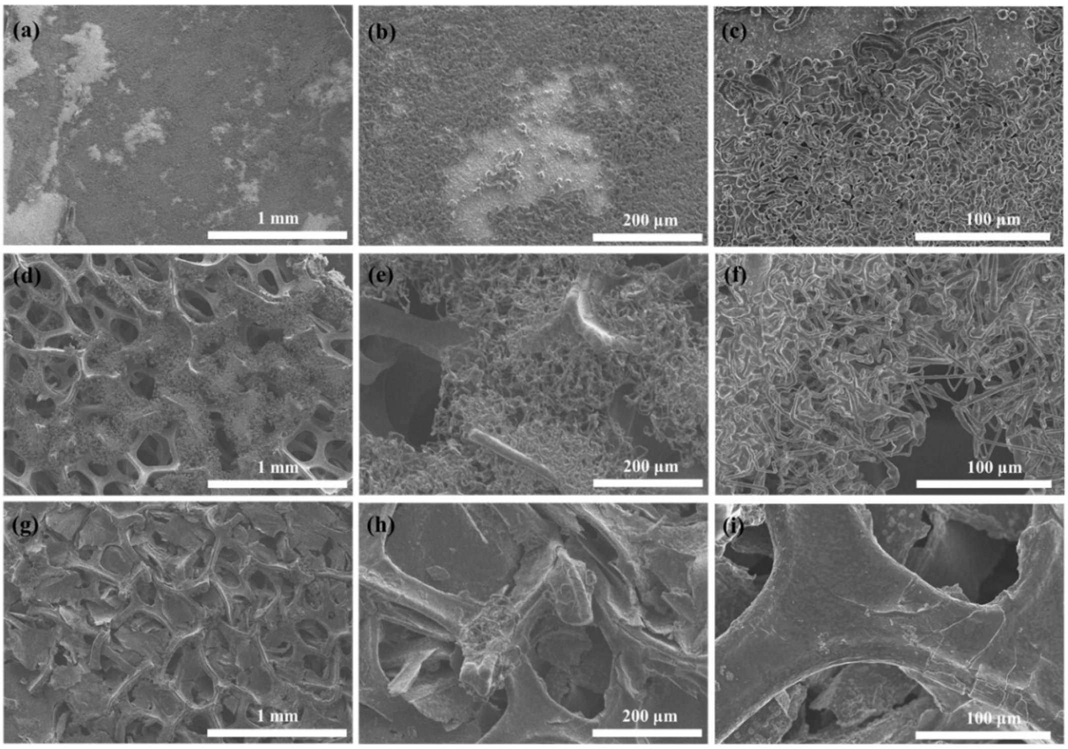

Fig. S3† SEM images of 1.0 mAh cm^{-2} of Li deposited at 0.5 mA cm^{-2} for the first cycle on a-c) bare Cu electrode, d-f) NF electrode, and g-i) 3D NGNF electrode.

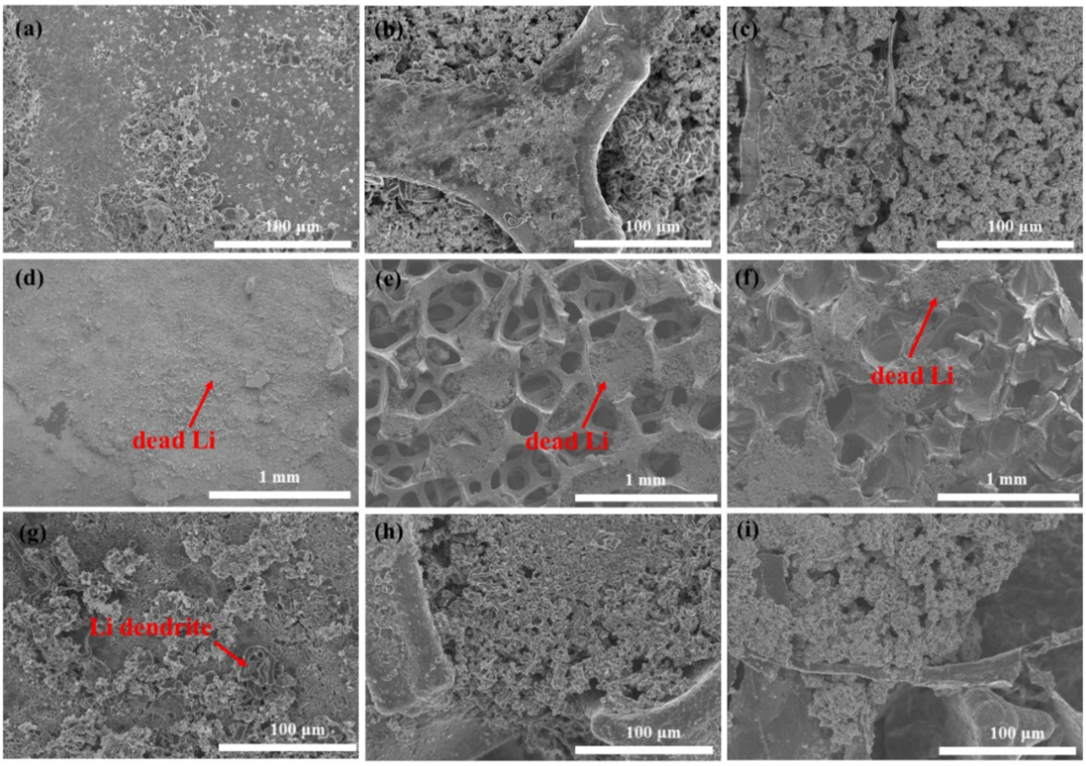

Fig. S4† SEM images of 1.0 mAh cm^{-2} of Li deposited at 1.0 mA cm^{-2} current density on a) bare Cu electrode, b) NF electrode, and c) NGNF electrode after the 50^{th} Li plating. The low magnification and high magnification images of d, g) Cu electrode, e, h) NF electrode, and f, i) NGNF electrode after the 50^{th} Li stripping.

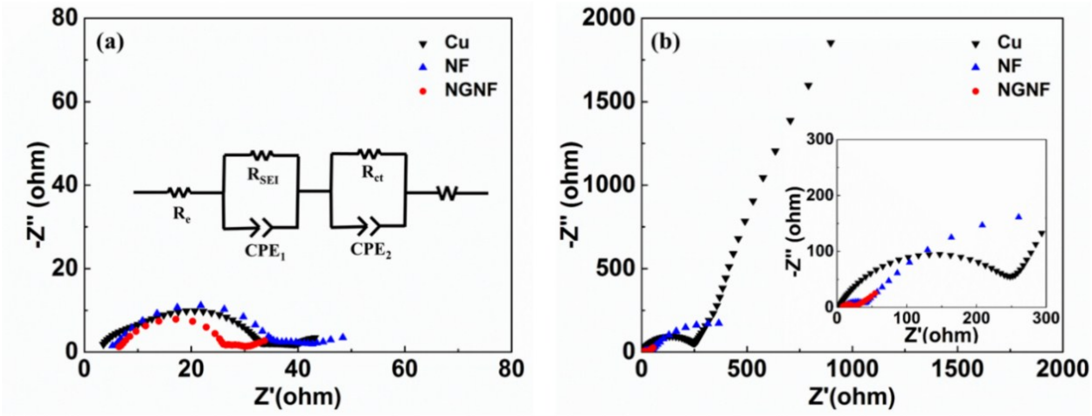

Fig. S5† Nyquist plots of the various half cells after the 1st a) plating and b) stripping. The inset of a) is the equivalent circuit used for analysing the results, and the inset of b) is an enlargement of the indicated region. R_e, R_{SEI}, and R_{ct} are the electrolyte resistance, SEI resistance, and charge transfer resistance, respectively. W is the Warburg impedance, and CPE_1 and CPE_2 are constant phase elements reflecting double layer capacitance.

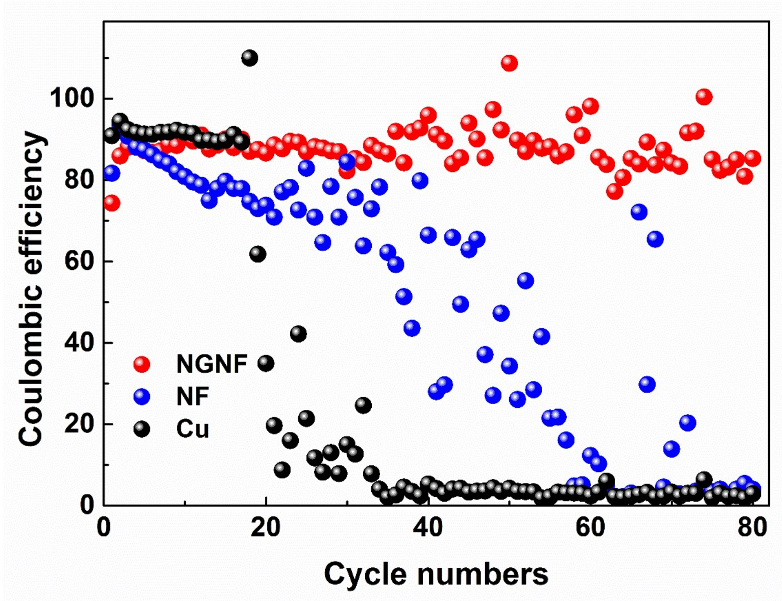

Fig. S6† Coulombic efficiency of bare Cu electrode, NF electrode, and NGNF electrode using carbonates-based electrolyte at 1.0 mA cm^{-2} with a cycling capacity of 1.0 mAh cm^{-2}.

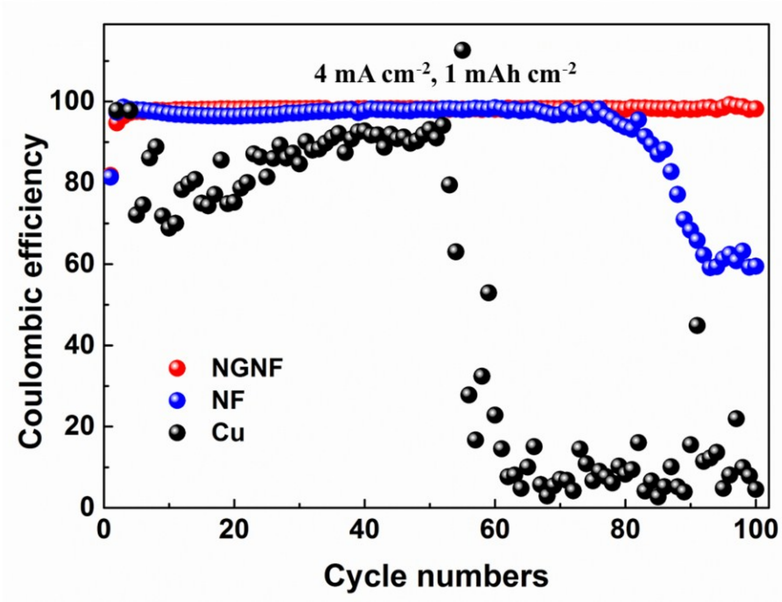

Fig. S7† Coulombic efficiency of bare Cu electrode, NF electrode, and NGNF electrode at 4.0 mA cm^{-2} with a cycling capacity of 1.0 mAh cm^{-2}.

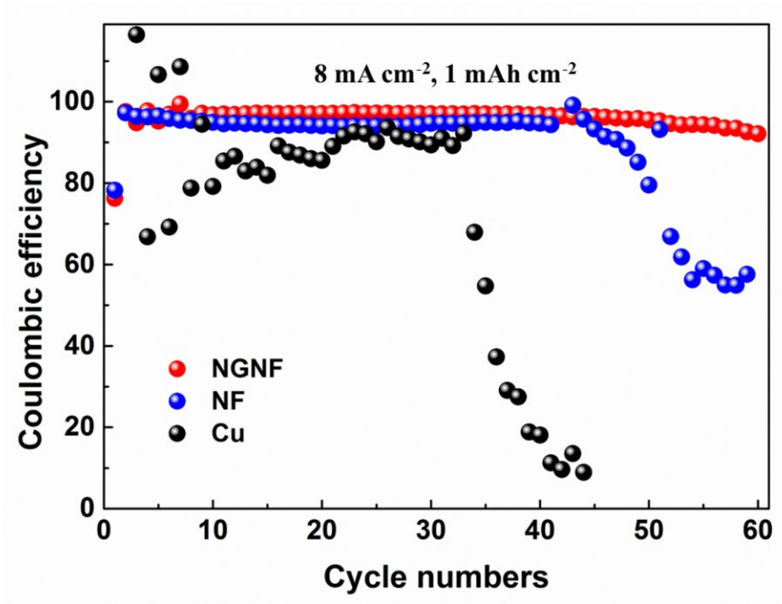

Fig. S8† Coulombic efficiency of bare Cu electrode, NF electrode, and NGNF electrode at 8.0 mA cm^{-2} with a cycling capacity of 1.0 mAh cm^{-2}.

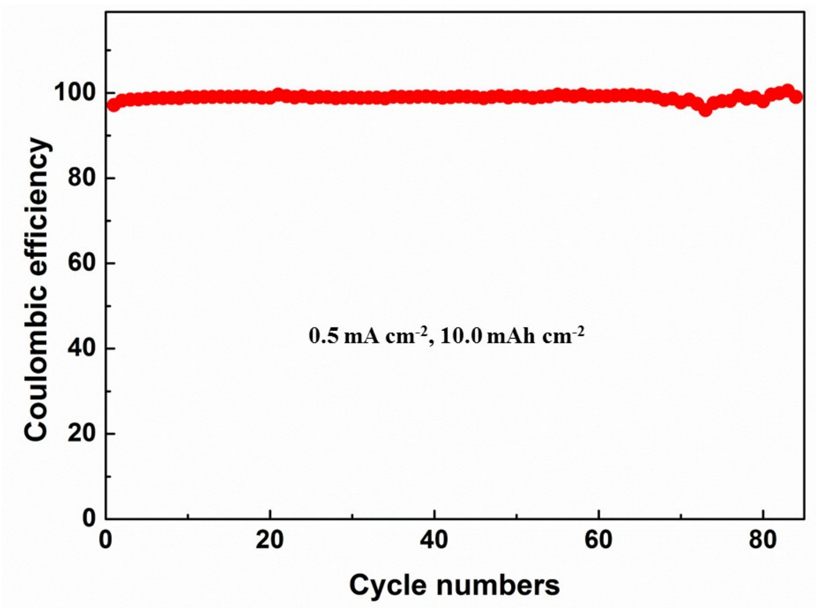

Fig. S9† Coulombic efficiency of NGNF electrode with a cycling capacity of 10.0 mAh cm^{-2} at a 0.5 mA cm^{-2}.

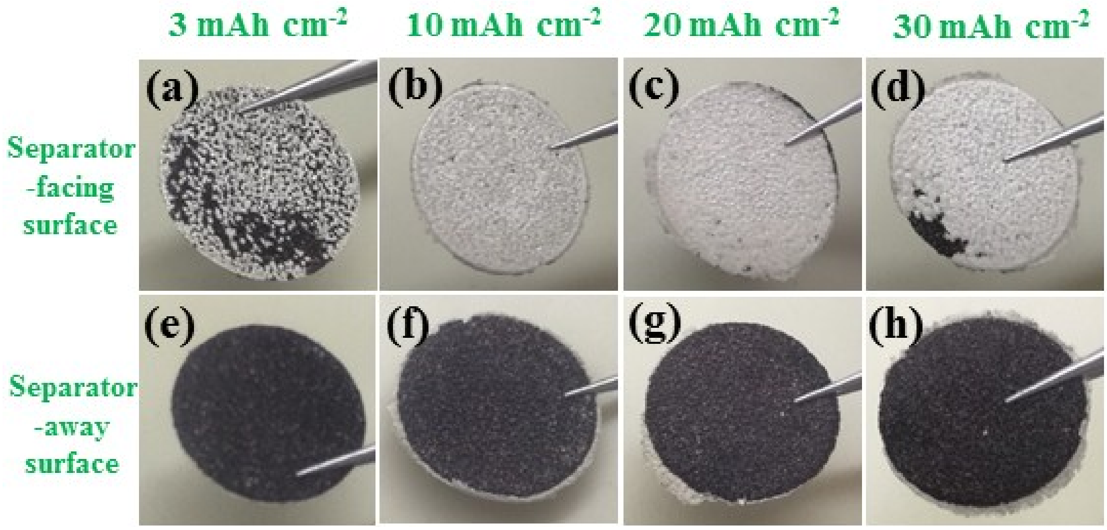

Fig. 10† a-d) Separator-facing surface and e-h) separator-away surface of Li-NGNF electrode with a capacity of 3 mAh cm^{-2}, 10 mAh cm^{-2}, 20 mAh cm^{-2}, and 30 mAh cm^{-2}. It can be seen that 10 mAh cm^{-2} Li deposition is the most suitable capacity for Li-NGNF electrode. This is because all the separator-facing surface areas of electrode have been covered by Li metal uniformly while the Li deposition capacity is the smallest among them.

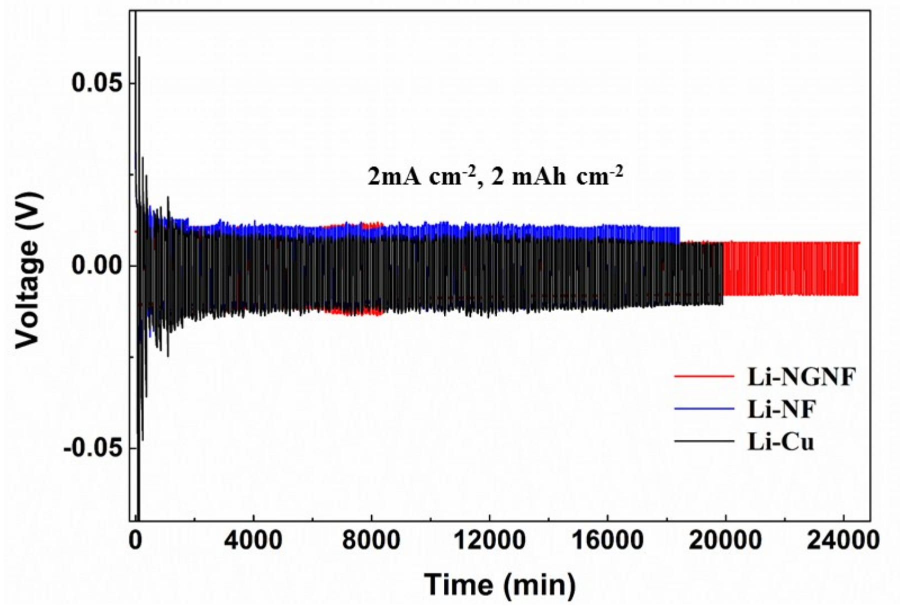

Fig. S11† Galvanostatic plating/stripping profiles of symmetric cells with NGNF, NF and Cu electrode at a current density of 2 mA cm^{-2}.

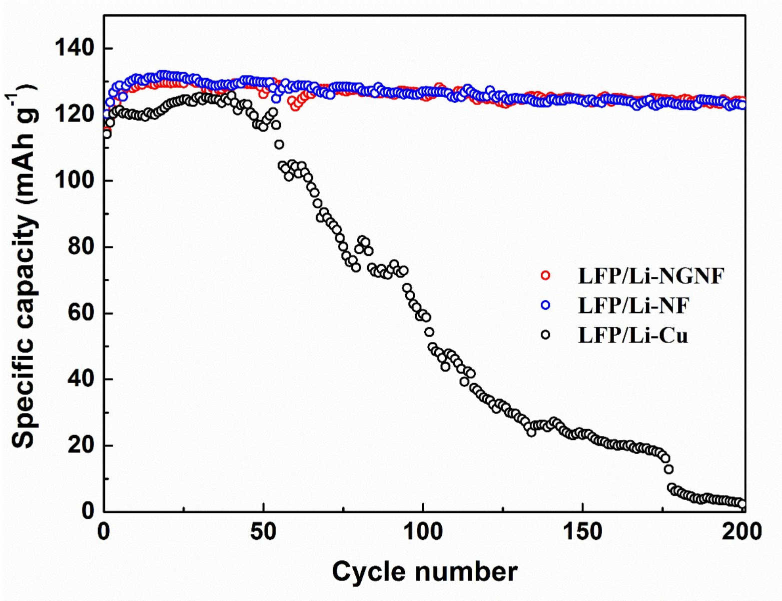

Fig. S12† Cycling performances of Li-Cu, Li-NF, and Li-NGNF anode in full cells with LFP cathode at 0.5 mA cm^{-2} (based on Li metal anode).

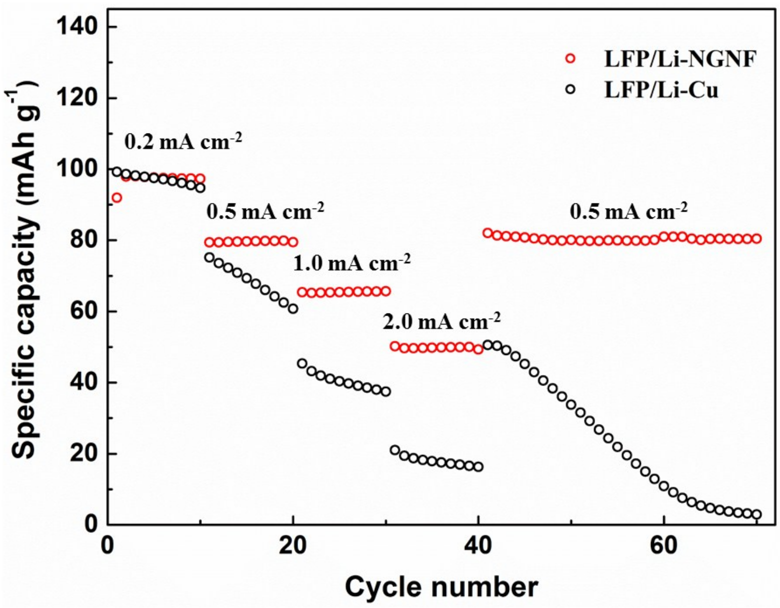

Fig. S13† Rate performance of LFP full cells with Li-NGNF and Li-Cu anodes at low temperature (-10 °C).

Table S1† Comparison of cycling performance achieved in this work with the previous reported anode substrates for Li metal batteries.

Ref.	**Anode substrates**	**Electrolytes**	**Current density ($mA\ cm^{-2}$)**	**Area capacity ($mAh\ cm^{-2}$)**	**Cycle numbers**	**Retention**
1	Hollow carbon nanospheres	1M LiTFSI in DOL/DME with 1% $LiNO_3$ and 100mM Li_2S_8 additives	0.5	1	150	98%
			1	1	150	97.5%
2	Graphene@Ni Scaffold	1M LiTFSI in DOL/DME with 2% $LiNO_3$	0.5	1	100	98%
			1	1	100	92%
3	NG coated Cu foil	1M LiTFSI in DOL/DME with 5% $LiNO_3$	1	1	< 200	98%
			2	1	100	96%
4	CuO Nanosheets	1M LiTFSI in DOL/DME with 1% $LiNO_3$	0.5	1	180	94%
			1	1	180	94%
5	Graphene flake	0.75M LiTFSI in DOL and 1.5M LiFSI in DME	2	1	50	93%
6	GF modifi ed Cu substrates	1M LiTFSI in DOL/DME with 2% $LiNO_3$	0.5	0.5	90	98%
			1	0.5	70	97%
			2	0.5	65	96%
7	PIANC coating on Stainless steel	1M LiTFSI in DOL/DME with 1% $LiNO_3$	1	0.5	240	97.6%
			2	0.5	150	92.9%
8	Silly putty modified electrode	1M LiTFSI in DOL/DME with 1% $LiNO_3$	0.5	1	120	97.6%
			1	1	120	97.0%
9	Co/Co4N-NC electrode	1M LiTFSI in DOL/DME with 1% $LiNO_3$	0.5	1	300	98.5%
			1	1	95	97.5%
			2	1	95	96.9%
This work	N-doped graphene modified nickel foam	1M LiTFSI in DOL/DME with 2% $LiNO_3$	**0.5**	**1**	**200**	**99.0%**
			1	**1**	**200**	**98.0%**
			2	**1**	**200**	**98.3%**

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