Electronic Supplementary Material (ESI) for ChemComm.

Electronic Supplementary Information:

Ladderlike carbon nanoarrays on 3D conducting skeletons enable uniform lithium nucleation for stable lithium metal anodes

Lin Liu, Ya-Xia Yin, Jin-Yi Li, Yu-Guo Guo, and Li-Jun Wan**

L. Liu, Dr. Y.-X. Yin, J.-Y. Li, Prof. Y.-G. Guo, Prof. L.-J. Wan
CAS Key Laboratory of Molecular Nanostructure and Nanotechnology CAS Research/Education Center for Excellence in Molecular Sciences, Institute of Chemistry, Chinese Academy of Sciences (CAS), Beijing 100190, P. R. China
E-mail: ygguo@iccas.ac.cn; wanlijun@iccas.ac.cn

L. Liu, Dr. Y.-X. Yin, J.-Y. Li, Prof. Y.-G. Guo, Prof. L.-J. Wan
University of Chinese Academy of Sciences (CAS), Beijing 100049, P. R. China

Experimental Section

Materials Synthesis

Preparation of the ZIF-8@Ni foam: The Ni foam was first pretreated with HCl solution containing polyvinyl pyrrolidone (PVP) to remove surface impurities and increase the affinity of the surface. After that, the Ni foam was washed with water and methanol for use. Then, the Ni foam was immersed in the metanol solution containing $Zn(NO_3)_2$ and 2-methylimidazole at proper ratio without disturbance for about 12 h.

Preparation of the LCNM@Ni and CM@Ni: The obtained ZIF-8@Ni at different ratio of reactants and reaction conditions was calcinated at 1100℃ for 3 h in Ar atmosphere. After that, the sample was washed with HCl solution and water, followed by drying. Then the LCNM@Ni and CM@Ni was punched out into circular disks (10 mm) as the electrode.

Materials Characterizations

The SEM images were acquired on a field emission scanning electron microscope (SU-8020). In order to observe the surface morphology of Li after cycling, the electrodes were disassembled from the coin cell in the glovebox followed by gentle rinse in DOL to remove residual electrolyte. Then a specially designed device was used to transfer the electrodes from the glove box to the vacuum chamber for ex situ SEM. X-ray diffraction (XRD) patterns of the carbon membrane were obtained on an X-ray diffractometer (Rigaku D/max-2500) with Cu Kα radiation (λ = 1.5406 Å).

Electrochemical Measurements

CR2032-type coin cells were assembled in an argon-filled glove box with the LCNM@Ni or Ni foam (control group) as the working electrode, Li foil as the

counter/reference electrode to evaluate the electrochemical deposition behavior and the Coulombic efficiency. 1 M lithium bis(trifluoromethanesulfonyl)imide in 1,3-dioxolane (DOL)/1,2-dimethoxyethane (DME) (1:1 w/w) with 1 wt % lithium nitrate was used as electrolyte. The Arbin BT2000 system was used to test the Coulombic efficiency at a constant current.

To probe the cycling stability and overpotential, 4 mA h cm^{-2} Li was first predeposited on the LCNM@Ni and Ni foam into half cells. Then LCNM@Ni-Li and Ni foam-Li electrodes were extracted from the half cells to assemble symmetrical cells. The Arbin BT2000 system was used to test the galvanostatic cycling. The potential was recorded over time at a constant current. Electrochemical impedance spectra measurement was conducted using a Princeton PARSTAT MC 1000 multi-channel electrochemical workstation with the frequency range of 100 kHz to 100 mHz.

Full cells were assembled with the $LiFePO_4$ as the cathode and LNCM@Ni-Li (4 mA h cm^{-2}) as the anode. $LiFePO_4$ electrode was prepared by mixing $LiFePO_4$, Super P, polyvinylidene difluoride (PVDF, Alfa Aesar) in the ratio of 90:5:5 with N-methyl-2-pyrrolidone as the solvent. The loading mass of the active material is ~12.5 mg cm^{-2}.

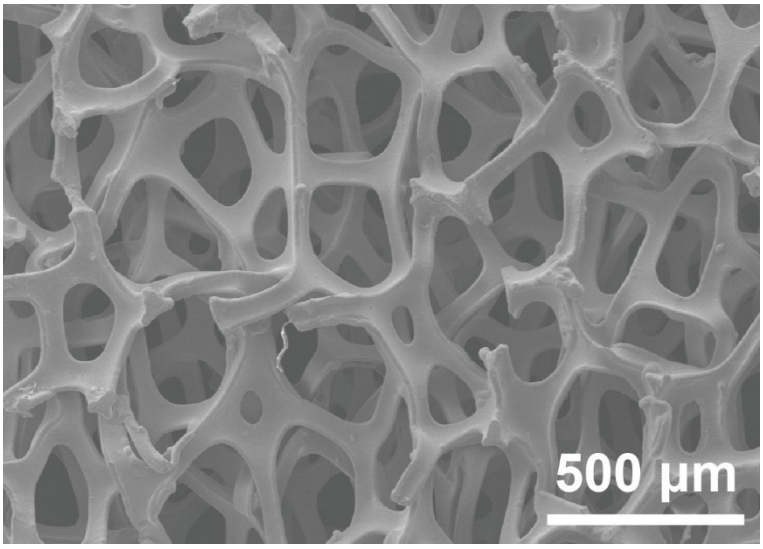

Fig. S1 SEM image of the Ni foam.

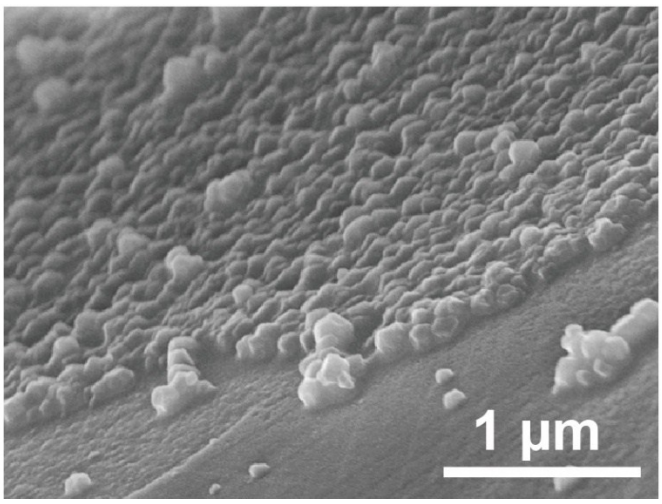

Fig. S2 Side view SEM image of the ZIF-8@Ni.

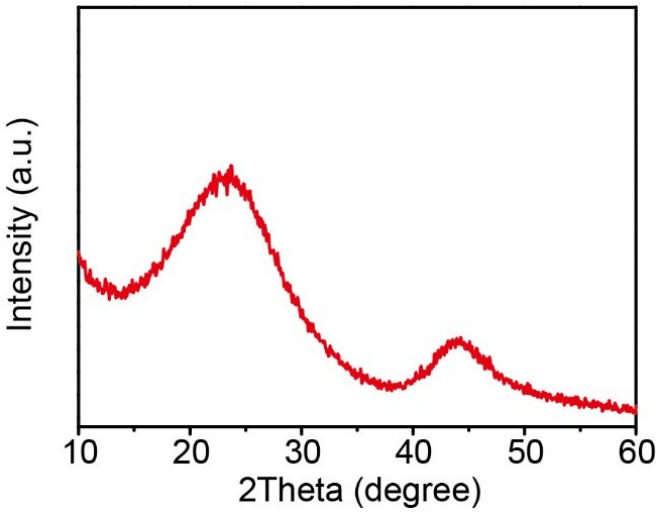

Fig. S3 XRD patterns of the ladderlike carbon membrane.

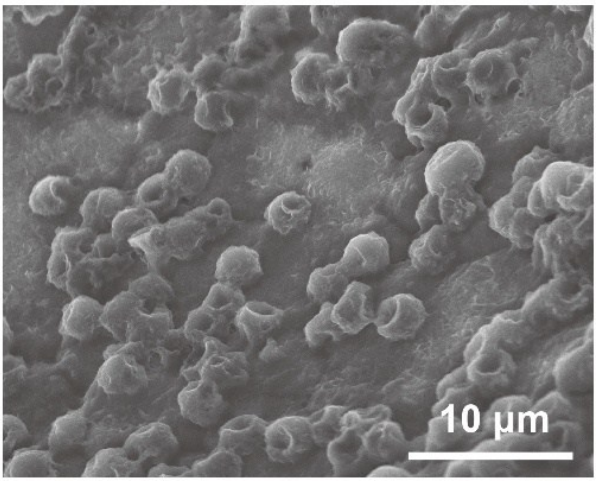

Fig. S4 SEM image of the LCNM@Ni-Li anode after 50 cycles at the current density of 2 mA cm^{-2} and the deposition areal capacity of 4 mA h cm^{-2}.

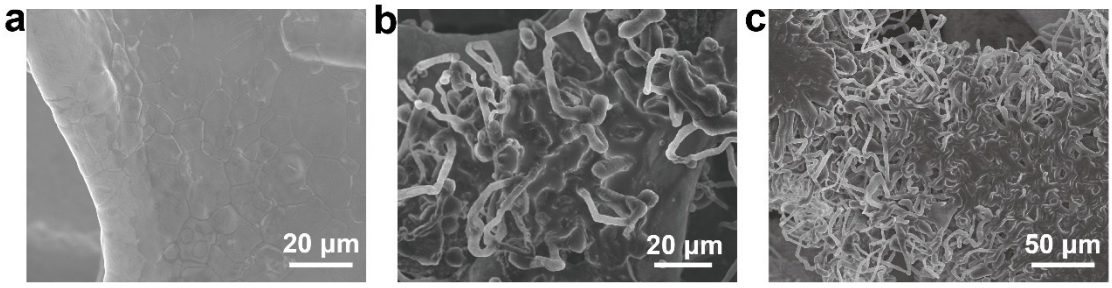

Fig. S5 SEM images of a) the pristine Ni foam without Li plating and after plating b) 2 mA h cm^{-2}, and c) 4 mA h cm^{-2} of Li metal on the Ni foam.

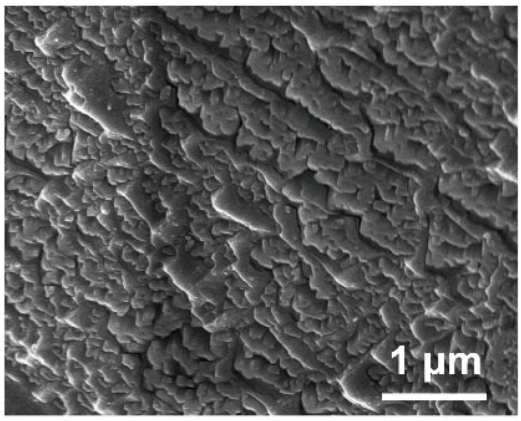

Fig. S6 SEM image of the CM@Ni.

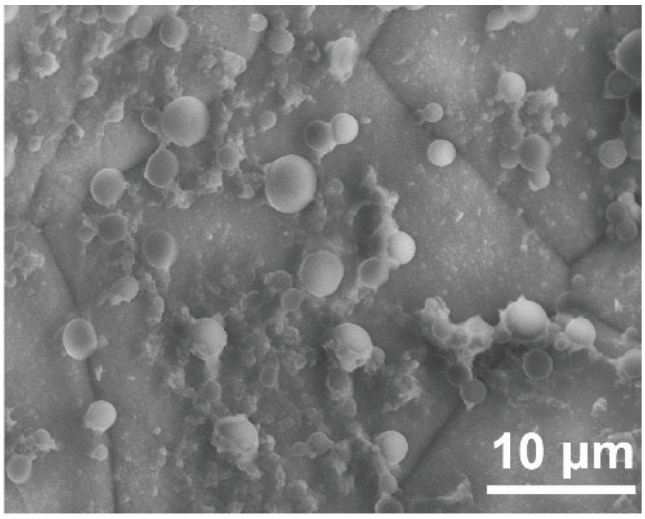

Fig. S7 SEM image of the CM@Ni after plating 2 mA h cm^{-2} of Li on it.

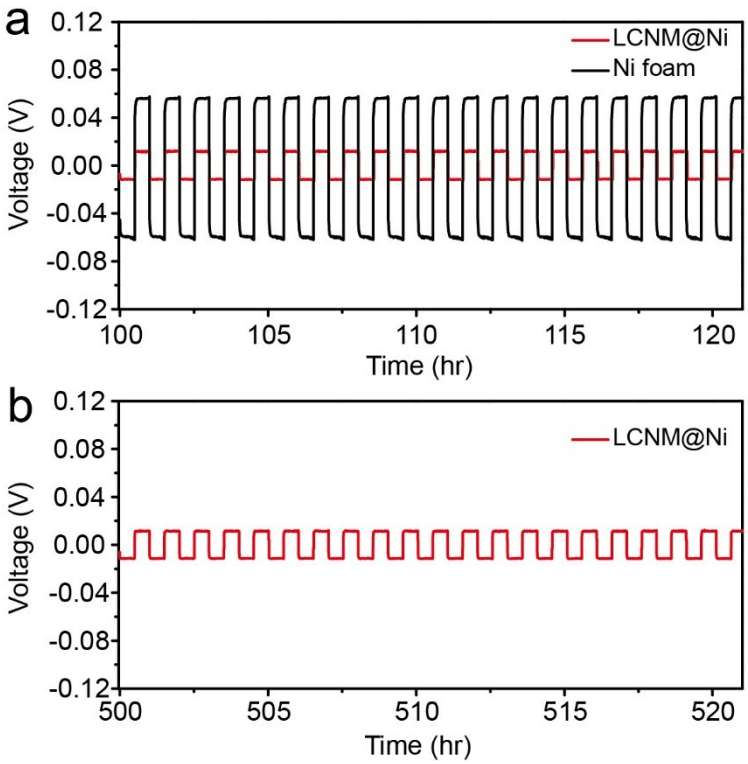

Fig. S8 The detailed voltage profiles from a) the 100th to the 120th cycle and b) the 500th to the 520th cycle at 2 mA cm^{-2} for 1 mA h cm^{-2}.

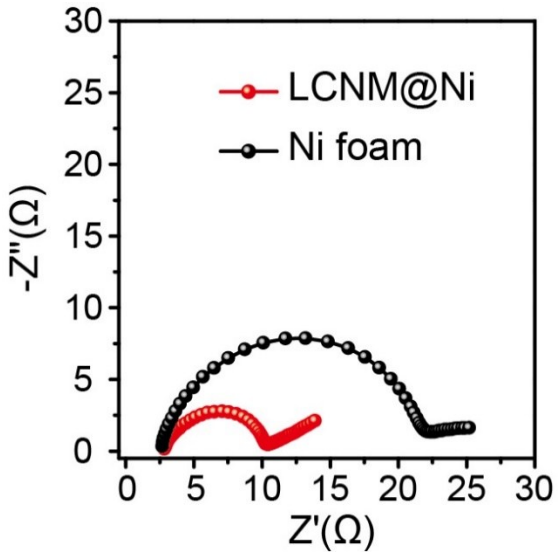

Fig. S9 Nyquist plot of the impedance spectra of the symmetric cells before cycling at 2 mA cm^{-2}.