Supplementary Information

Densely Vertical-Grown NiFe Hydroxide Nanosheets on a 3D Nickel Skeleton as a Dentrite-free Lithium Anode

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Experimental Section

Chemicals

Nickel foam (1.5 mm thickness) was purchased from Sai Bo electrochemical materials (China). Ferric nitrides, urea ($CO(NH_2)_2$) and ethylene glycol were purchased from Chengdu Kelong Chemical Reagent Factory (China). Ammonium fluoride (NH_4F) was purchased from Chongqing Chuandong Chemical Group Co., Ltd. Li-S electrolyte (LS-002, 1.0 M LiTFSI in DME: DOL = 1:1 vol% with 1.0% $LiNO_3$) was purchased from www.DoDoChem.com. PVDF, N-methyl-2-pyrrolidone (NMP), and conductive carbon black (graphite&carbon super P) were purchased from Hefei Kejing Material Technology Co., Ltd.

Characterization

The morphology of samples was investigated through scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The composition of samples was studied by scanning TEM (STEM) and powder X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS) energy-dispersive X-ray spectroscopy (EDX).

Electrochemical measurements

Electrochemical measurements were mainly conducted through the LAND electrochemical testing system. CR 2025 coin cells were assembled to evaluate the symmetric cell property (galvanostatic cycling performance). CR 2032 coin cells were assembled to conduct the full cell test ($LiFePO_4$ full cell test).

Synthesis of DVS-NFOH@NF and SVS-NFOH@NF

DVS-NFOH@NF was synthesized through a one-step solvothermal method. Nickel foam (NF, 0.15×2×3 cm^3) was sonicated in 3 M HCl solution for 30 min and washed with de-ionized water three times. Then, the NF was sonicated in ethanol for 10 min to remove the greasy dirt. Meanwhile, 3.0 mM $Fe(NO_3)_3$, 7.9 mM NH_4F and 9.9 mM urea were dissolved in 30 ml solvent (**10 ml deionized water and 20 ml ethylene glycol**) and **stirred for 20 min**. Subsequently, the as-obtained solution was added into a 50 ml Teflon-lined stainless steel autoclave, followed by the immersion in clean nickel foam. Then, the autoclave was sealed and heated up to 120 ℃ for 12 h. The resultant in pale green was sonicated three times in deionized water for 5 min each time and dried at 60 ℃ for 8 h, then the DVS-NFOH@NF can be obtained. The SVS-NFOH@NF was obtained in the same condition, except for **changing the solvent to 25 ml deionized water and 5 ml ethylene glycol**.

Synthesis of Li/DVS-NFOH@NF, Li/SVS-NFOH@NF and Li@NF

The as-obtained DVS-NFOH@NF/SVS-NFOH@NF was punched to round shape with 14 mm diameter, then served as the cathode of coin cell to assemble with lithium foil anode (CR 2025 coin cell shell, 100 μL **LS-002** Li-S electrolyte and 0.2 mm stainless steel disc). Subsequently, lithium was deposited to DVS-NFOH@NF/ SVS-NFOH@NF using galvanostatic method (**discharge at 2 mA cm^{-2} current density for 2, 4, 6, 8 h**) and the Li/DVS-NFOH@NF/Li/SVS-NFOH@NF was obtained. The Li@NF was obtained in the same condition, except for replacing the cathode with nickel foam.

Preparation of the $LiFePO_4$ cathode

80 mg commercial $LiFePO_4$ powder, 10 mg conductive carbon black (graphite&carbon super P) and 10 mg PVDF were mixed in a mortar and milled for 1 h. Then, 600 μL N-methyl-2-pyrrolidone (NMP) were added as the solvent for PVDF and milled for 0.5 h to **obtain homogenous ink**. The mixture was then transferred to a clean Al foil (5×10 cm^2), which was attached to a smooth glass plate. Followed by a knife coating process with the proper scraper (450 μm). After that, the product was dried in an air-dry oven and a vacuum dry oven for both 8 h under 40 ℃.

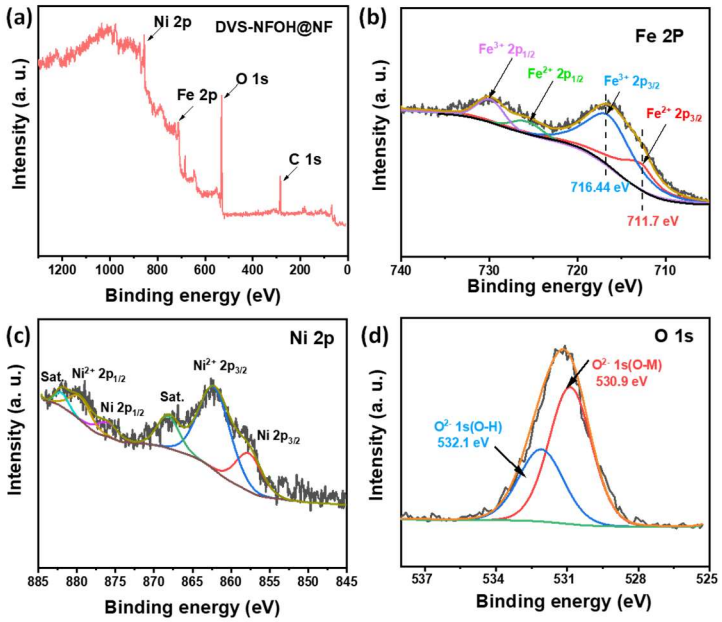

Figure S1. (a) X-ray photoelectron spectroscopy survey spectrum of the DVS-NFOH@NF and the corresponding XPS spectra for (b) Fe 2p, (c) Ni 2p and (d) O 1s.

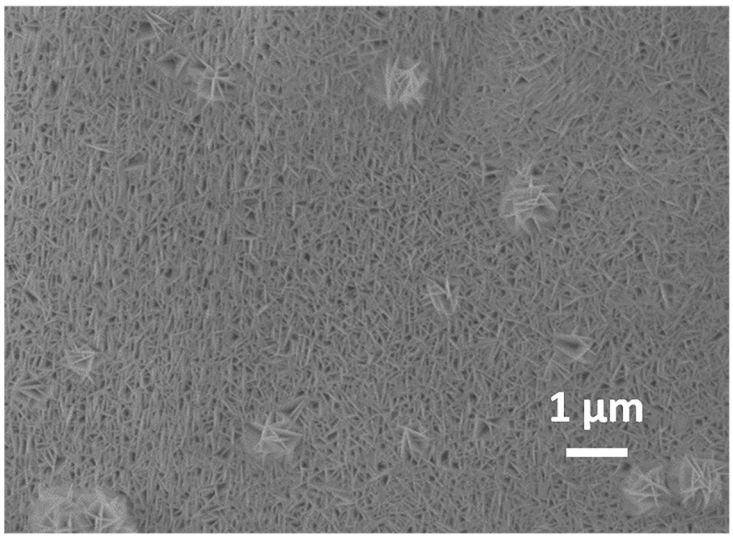

Figure S2. SEM images of DVS-NFOH@NF.

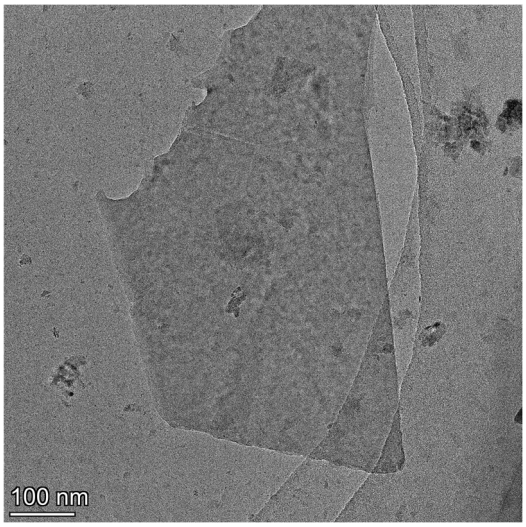

Figure S3. TEM images of DVS-NFOH@NF.

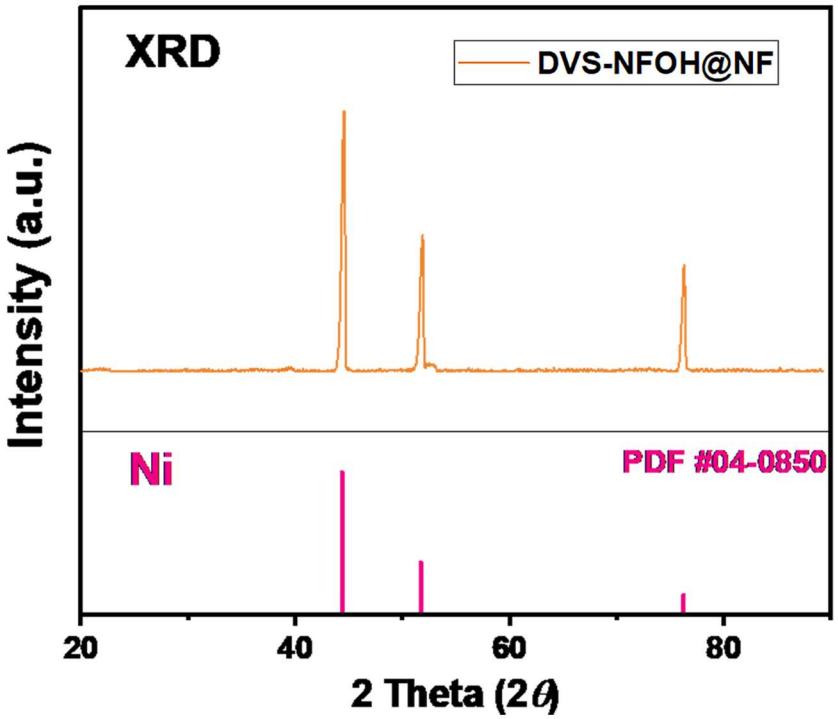

Figure S4. XRD patterns of DVS-NFOH@NF.

The XRD results only shows the typical peaks of nickel which suggests the $NiFe(OH)_x$ may exist in the form of amorphous state.

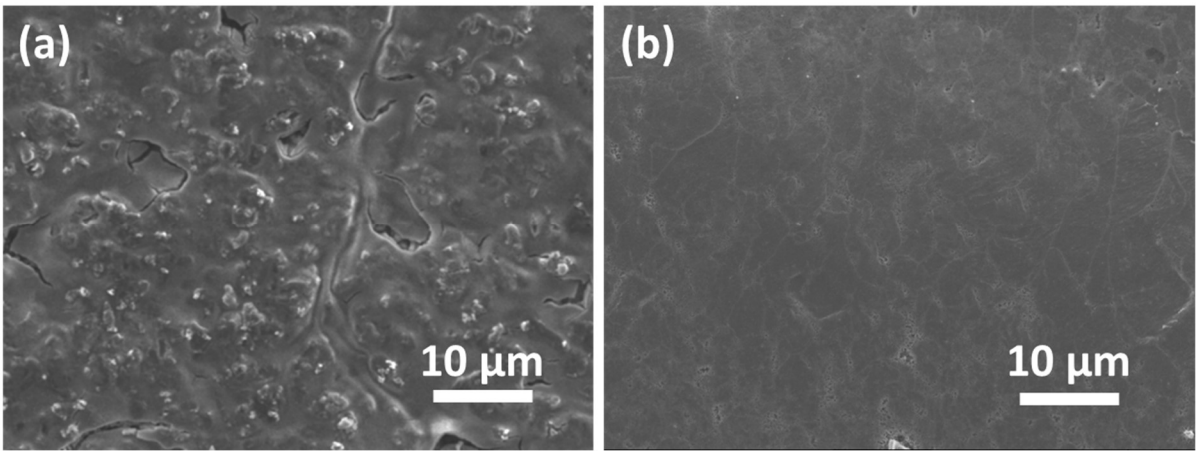

Figure S5. SEM images of lithium deposition for 8 h under 2 mA cm^{-2} on (a) SVS-NFOH@NF and (b) DVS-NFOH@NF.

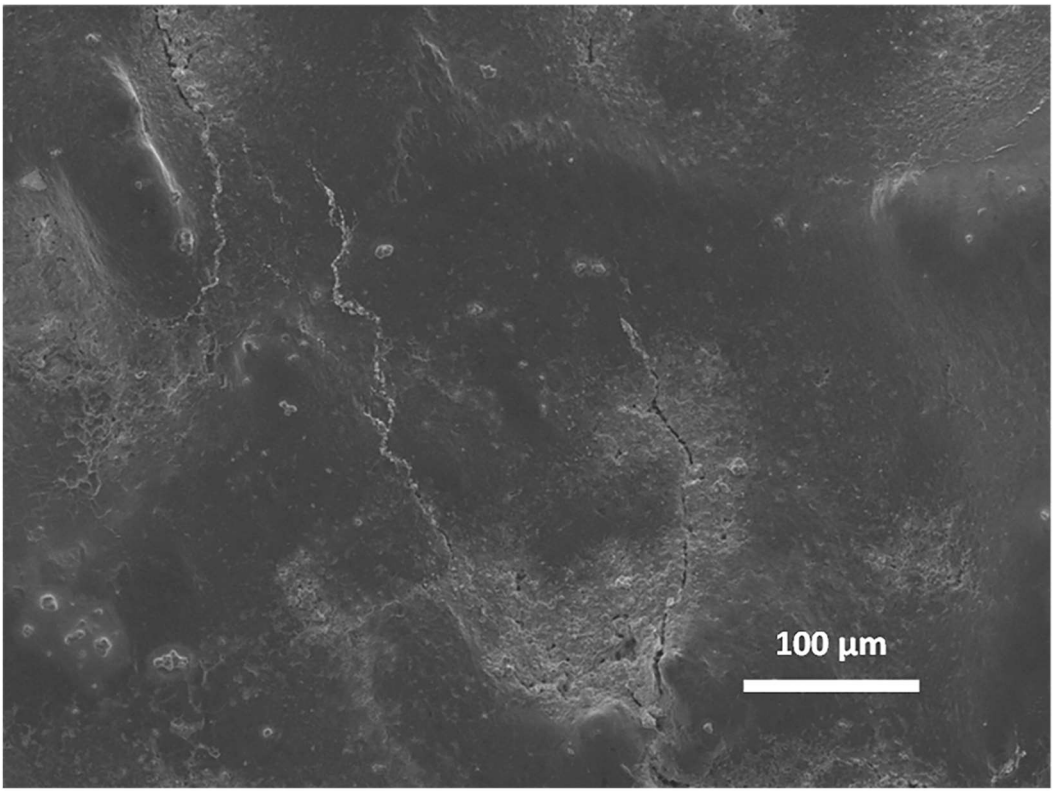

Figure S6. SEM images of Li/DVS-NFOH@NF deposited at 2 mA cm^{-2} for 8h.

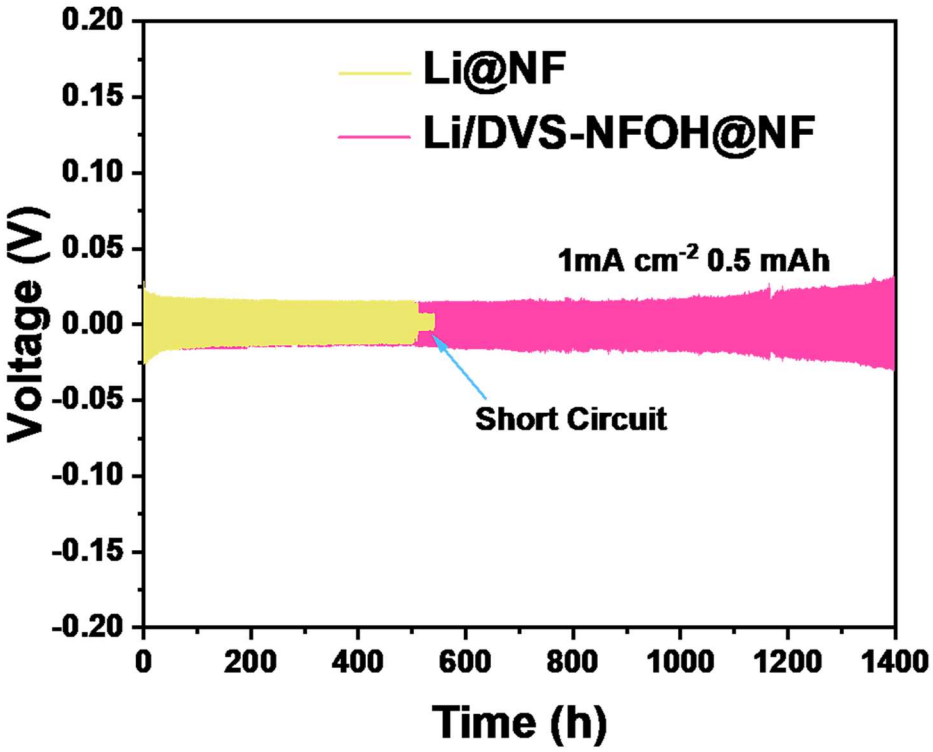

Figure S7. Symmetrical cell tests of Li@NF and Li/DVS-NFOH@NF at 1 mA cm^{-2} current density for 0.5 h.

Table S1. Recent researches on Li metal anode and corresponding performace.

Reference	Materials	Electrolyte	Condition	Over potential (mV)	Time (h)
This work	DVS-NFOH@NF	1.0 M LiTFSI in DME/DOL (1:1) + 1 wt% $LiNO_3$	3 mA cm^{-2} 1 mAh cm^{-2}	25	1000 (cycle)
			1 mA cm^{-2} 0.5 mAh cm^{-2}	14	1400
ACS Energy Lett. 2020, 5, 3108	3D porous nickel	LiFSI in DMC	2 mA cm^{-2} 1 mAh cm^{-2}	200	250
Adv. Mater. 2018, 30, 1802156.	3D graphene foam	1 M LiTFSI in DOL/DME (1:1) + 2 wt% $LiNO_3$	1 mA cm^{-2} 1 mAh cm^{-2}	25	320
Energy Storage Mater. 2020, 29, 332.	N-doped CNTs@Ni Foam	1 M LiTFSI in DOL/DME (1:1) + 1 wt% $LiNO_3$	3 mA cm^{-2} 1 mAh cm^{-2}	88	500 (Cycle)
Adv. Energy Mater. 2019, 9, 1803186.	g-C_3N_4@Ni foam	1 M LiTFSI in DOL/DME (1:1) + 1 wt% $LiNO_3$	1 mA cm^{-2} 1 mAh cm^{-2}	15	900
Nano Energy 2019, 60, 257.	Cu_xO@carbon felt	1 M $LiPF_6$ in EC/DEC (1:1)	1 mA cm^{-2} 1 mAh cm^{-2}	30	1000
Adv. Funct. Mater. 2019, 29, 1808847.	Co_3O_4 nanofber@ carbon sheet	1 M LiTFSI in DOL/DME (1:1)	1 mA cm^{-2} 1 mAh cm^{-2}	50	800
ACS Nano 2019, 13, 8337.	Carbon nanosheet@ Cu foam	1 M LiTFSI in DOL/DME (1:1) + 2 wt% $LiNO_3$	0.5 mA cm^{-2} 1 mAh cm^{-2}	10	1600
Nano Energy 2020, 69, 104471.	Au@carbon fber paper	1 M LiTFSI in DOL/DME (1:1) + 2 wt% $LiNO_3$	1 mA cm^{-2} 1 mAh cm^{-2}	18	1000
Adv. Funct. Mater. 2020, 30, 1906444	Vertically aligned CNFs@Cu foil	1 M LiTFSI in DOL/DME (1:1) + 1 wt% $LiNO_3$	5 mA cm^{-2} 5 mAh cm^{-2}	200	550
Adv. Mater. 2019, 31, 1807131.	LiC_6 layers on carbon fiber	1 M LiTFSI in DOL/DME (1:1) + 5 wt% $LiNO_3$	1 mA cm^{-2} 1 mAh cm^{-2}	25	1000