

Supporting Information

Superhydrophilic “Nanoglue” Stabilizing the Metal Hydroxides onto Carbon Materials for High-Energy and Ultralong-Life Asymmetric Supercapacitors

Shaofeng Li,^a Chang Yu,^{*a} Juan Yang,^a Changtai Zhao,^a Mengdi Zhang,^a Huawei Huang,^a Zhibin Liu,^a Wei Guo,^a and Jieshan Qiu^{*a,b}

^aState Key Lab of Fine Chemicals, School of Chemical Engineering, Liaoning Key Lab for Energy Materials and Chemical Engineering, Dalian University of Technology, Dalian 116024, China. E-mail: chang.yu@dlut.edu.cn; jqu@dlut.edu.cn

^bSchool of Chemical Engineering and Technology, Xi'an Jiaotong University, Xi'an 710049, China. Email: jasonqiu@xjtu.edu.cn

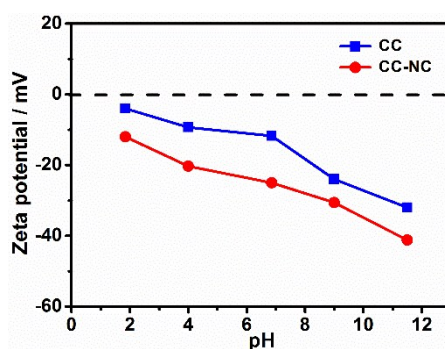


Fig. S1 Surface charge variation of the CC and CC-NC samples at different pH values by zeta potential measurements.

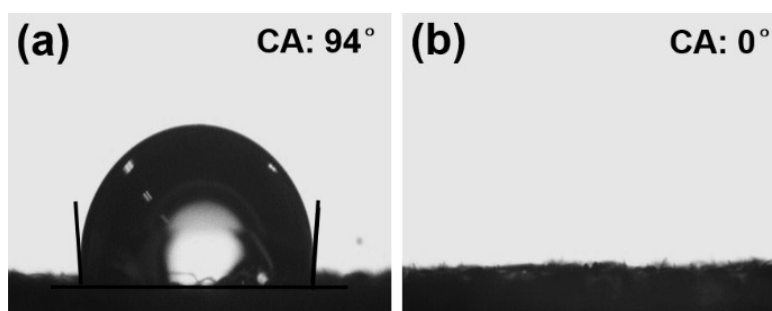


Fig. S2 The contact angle of (a) CC-LDH and (b) CC-NC-LDH hybrids.

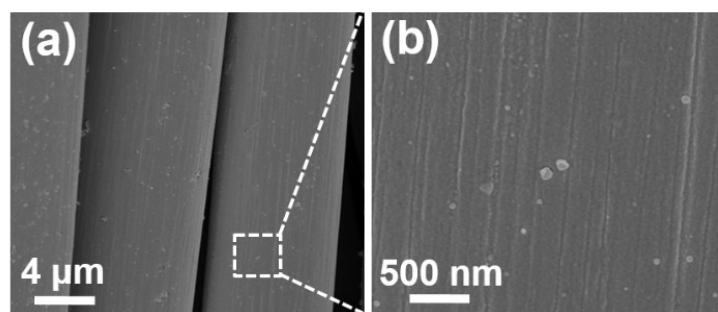


Fig. S3 The typical FE-SEM images of the CC-P sample.

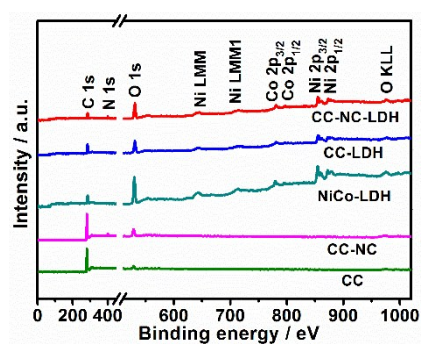


Fig. S4 XPS survey spectra of CC, CC-NC, NiCo-LDH, CC-LDH and CC-NC-LDH samples.

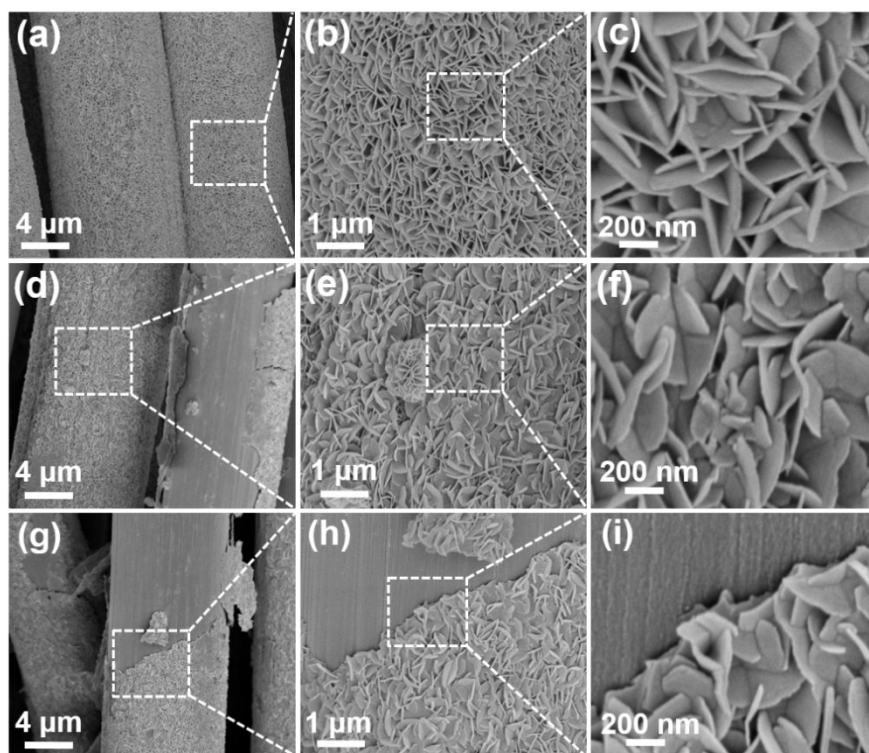


Fig. S5 The typical FE-SEM images of (a-c) the CC-NC-LDH hybrids after 30 min sonication treatment; (d-f) the CC-HNO₃-LDH without 30 min sonication treatment; and (g-i) the CC-HNO₃-LDH after 30 min sonication treatment.

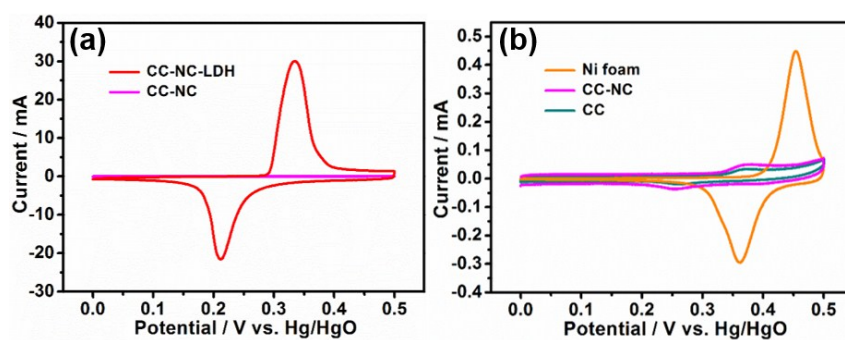


Fig. S6 CV curves of the pristine CC, pristine Ni foam, CC-NC and CC-NC-LDH hybrid electrodes in a potential range of 0 to 0.5 V at a scan rate of 10 mV s⁻¹.

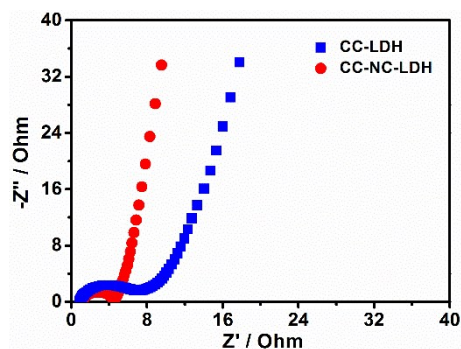


Fig. S7 Nyquist plots of the CC-LDH and CC-NC-LDH hybrid electrodes.

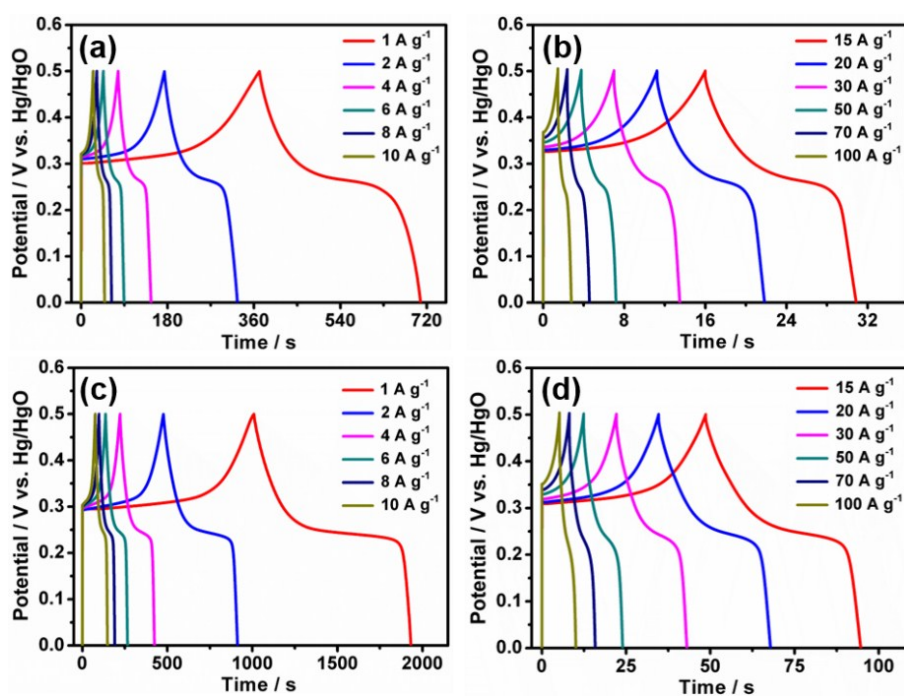


Fig. S8 Electrochemical performance of the samples measured in 6 M KOH electrolyte: charge/discharge curves of (a, b) CC-LDH and (c, d) CC-NC-LDH hybrid electrodes at different current densities ranging from 1 to 10 A g⁻¹ and 15 to 100 A g⁻¹.

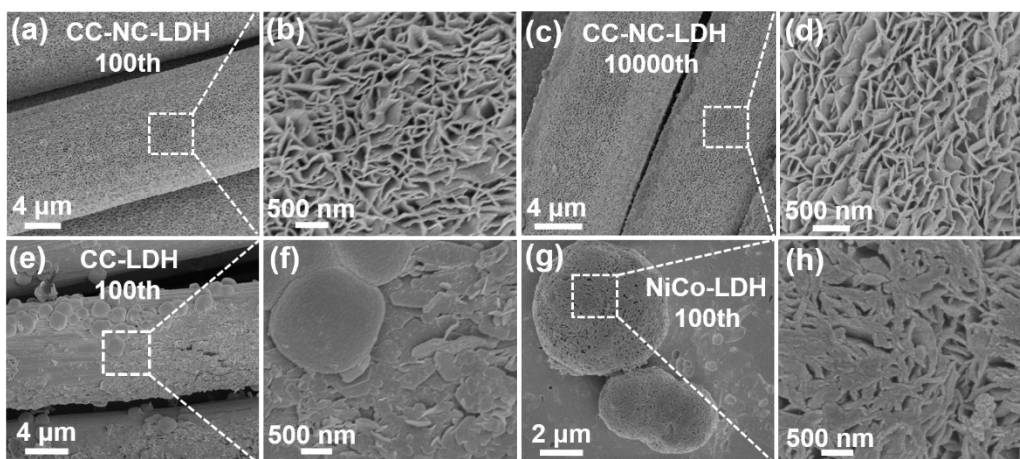


Fig. S9 Typical FE-SEM images of the CC-NC-LDH hybrids after (a, b) 100 cycles and (c, d) 10000 cycles at a current density of 10 A g^{-1} ; (e, f) CC-LDH and (g, h) pristine NiCo-LDH after 100 cycles at a current density of 10 A g^{-1} .

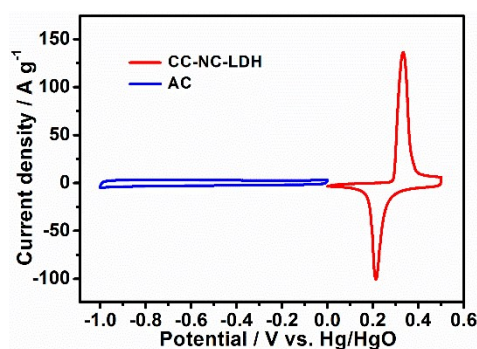


Fig. S10 CV curves of the AC and CC-NC-LDH hybrid electrodes at a scan rate of 10 mV s^{-1} .

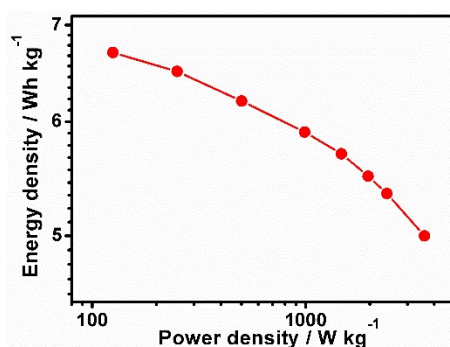


Fig. S11 Ragone plot related to energy and power density of the AC//AC symmetric supercapacitors, in which an energy density of ca. 6.7 Wh kg^{-1} is delivered at a power density of 125 W kg^{-1} , and the energy density of 5 Wh kg^{-1} is achieved at a power density of 3.6 kW kg^{-1} .

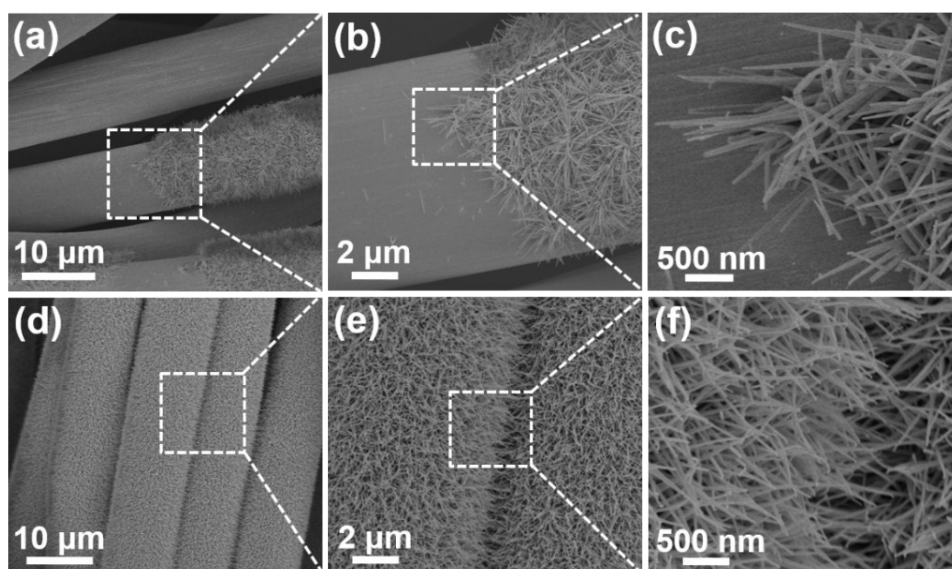


Fig. S12 Typical FE-SEM images of (a, b, c) the CC-CH and (d, e, f) CC-NC-CH hybrids.

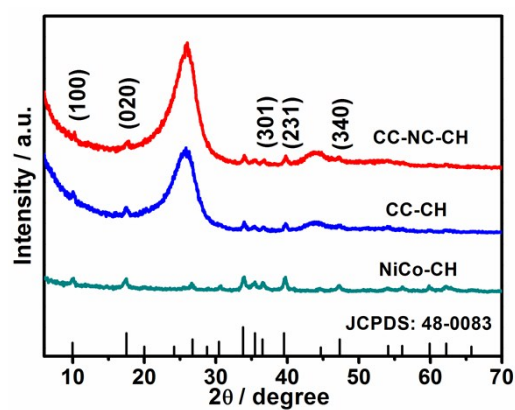


Fig. S13 Typical XRD patterns of the as-made CC-NC-CH, CC-CH and pristine NiCo-CH.

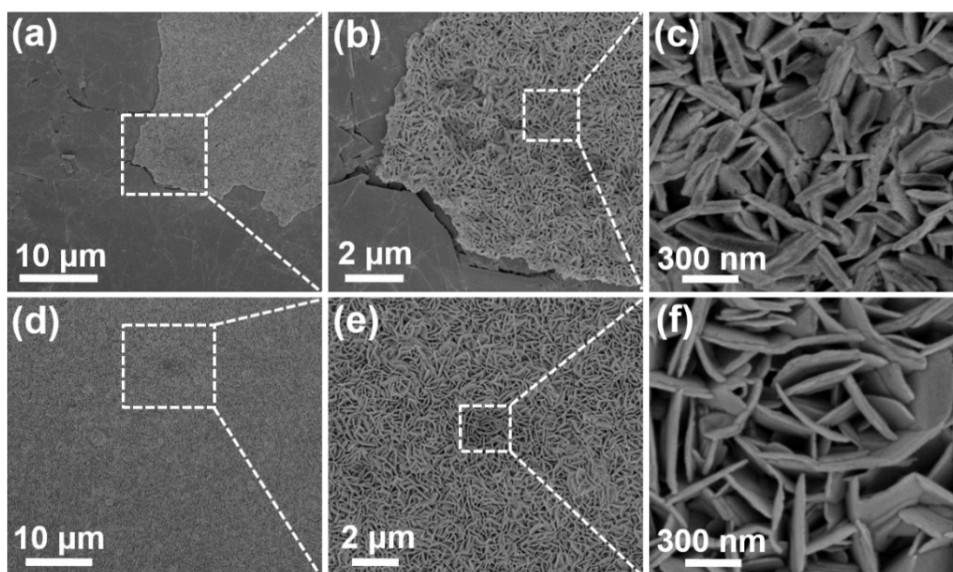


Fig. S14 Typical FE-SEM images of (a, b, c) the G-LDH and (d, e, f) G-NC-LDH nanohybrids.

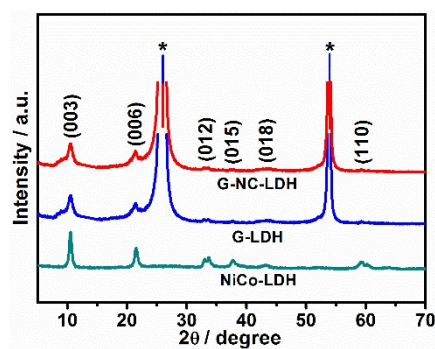


Fig. S15 Typical XRD patterns of the as-made G-NC-LDH, G-LDH and pristine NiCo-LDH.

Table S1 The relative contents of elements in CC-NC sample derived from XPS data.

CC-NC	C	O	N
at. %	87.19	11.74	1.07

Table S2 The Ni and Co contents in CC-NC-LDH, CC-LDH and NiCo-LDH samples analysed by ICP technique.

Sample	Ni (wt%)	Co (wt%)
CC-NC-LDH	1.501	0.499
CC-LDH	1.499	0.498
NiCo-LDH	1.503	0.501

Table S3 The atomic fractions of the nitrogen bonding states calculated from the XPS N 1s peaks of CC-NC and CC-NC-LDH nanohybrids.

Sample	pyrrolic-N (%)	graphitic-N (%)
CC-NC	78.2	21.8
CC-NC-LDH	70.9	29.1

Table S4 A comparison of electrochemical performance for CC-NC-LDH hybrid electrodes with some representative Ni-Co oxides or hydroxides in literature, all in electrolyte of 6 M KOH.

Sample	System voltage (V)	Electrochemical performance			reference
		Specific capacitance	Capacitance retention	Stability	
CC-NC-LDH	0.50 V vs. Hg/HgO	1817 F g ⁻¹ at 1 A g ⁻¹	60% at 100 A g ⁻¹	88% after 10000 cycles	This work
Co ₃ O ₄ /NiCo ₂ O ₄ double-shelled nanocages	0.42 V vs. SCE	972 F g ⁻¹ at 5 A g ⁻¹	63% at 50 A g ⁻¹	92% after 12000 cycles	[1]
CoAl-LDH/graphene	0.40 V vs. SCE	1043 F g ⁻¹ at 1 A g ⁻¹	87% at 20 A g ⁻¹	88% after 3000 cycles	[2]
NiCoAl-LDH /NiCo-CH nanowire	0.48 V vs. Hg/HgO	1297 F g ⁻¹ at 1 A g ⁻¹	59% at 30 A g ⁻¹	--	[3]
NiCo ₂ O ₄ @Au nanotubes	0.45 V vs. SCE	1390 F g ⁻¹ at 5 A g ⁻¹	38% at 100 A g ⁻¹	85% after 10000 cycles	[4]
NiCo ₂ O ₄ microtubes	0.53 V vs. SCE	1387.9 F g ⁻¹ at 2 A g ⁻¹	38% at 30 A g ⁻¹	89% after 12000 cycles	[5]
NiCo ₂ O ₄ hollow spheres	0.50 V vs. SCE	1141 F g ⁻¹ at 1 A g ⁻¹	69% at 15 A g ⁻¹	95% after 4000 cycles	[6]
aCF-NiCo-double hydroxide	0.65 V vs. Hg/HgO	1281 F g ⁻¹ at 0.5 A g ⁻¹	70% at 20 A g ⁻¹	--	[7]
NiCo-CH nanowire-Graphene-CNT	0.48 V vs. Hg/HgO	1146 F g ⁻¹ at 1 A g ⁻¹	40% at 100 A g ⁻¹	95% after 2500 cycles	[8]

a) 3E refers to three electrode test.

Reference

- [1] H. Hu, B. Y. Guan, B. Y. Xia and X. W. Lou, *J. Am. Chem. Soc.*, 2015, **137**, 5590-5595.
- [2] X. Wu , L. Jiang , C. Long , T. Wei and Z. Fan, *Adv. Funct. Mater.*, 2015, **25**, 1648-1655.
- [3] J. Yang , C. Yu , X. Fan and J. Qiu, *Adv. Energy Mater.*, 2014, **4**, 1400761.
- [4] J. Zhu, Z. Xu and B. Lu, *Nano Energy*, 2014, **7**, 114-123.
- [5] F. Ma, L. Yu, C. Xu and X. W. Lou, *Energy Environ. Sci.*, 2016, **9**, 862-866.
- [6] L. Shen, L. Yu, X. Y. Yu, X. Zhang and X. W. Lou, *Angew. Chem. Int. Ed.*, 2014, **54**, 1868-1872.

- [7] T. Li, W. Zhang, L. Zhi , H. Yu, L. Dang , F. Shi , H. Xu, F. Hu, Z. Liu, Z. Lei and J.Qiu, *Nano Energy*, 2016, **30**, 9-17.
- [8] J. Yang, C. Yu, X. Fan, C. Zhao and J. Qiu, *Adv. Funct. Mater.*, 2015, **25**, 2109-2116.