## **Supporting information**

Flexible All-solid-state Ultrahigh-energy Asymmetric Supercapacitors Based on Tailored Morphology of NiCoO<sub>2</sub>/Ni(OH)<sub>2</sub>/Co(OH)<sub>2</sub> Electrodes

Tianfeng Qin,<sup>a</sup> Haoqian Li,<sup>a</sup> Ruojia Ren,<sup>a</sup> Jiaxin Hao,<sup>a</sup> Yuxiang Wen,<sup>a</sup> Zilei Wang,<sup>a</sup> Juanjuan Huang,<sup>a</sup> Deyan He,<sup>a</sup> Guozhong Cao,<sup>\*a,b</sup> Shanglong Peng<sup>\*a</sup>

<sup>a</sup> Key Laboratory for Magnetism and Magnetic Materials of the Ministry of Education, School of Physical Science and Technology, Lanzhou University, Lanzhou 730000, P.R. China
 <sup>b</sup> Department of Materials and Engineering, University of Washington, Seattle, Washington 98195-2120, United States

\* E-mail address: <a href="mailto:pengshl@lzu.edu.cn">pengshl@lzu.edu.cn</a>;

E-mail address: gzcao@u.washington.edu; Tel.: +1 206 616 9084



Fig. S1 (a) High and (c) low-magnification SEM images for Ni sample. (c) Enlarged-view SEM image of CoNi sample.



Fig. S2 (a) XRD pattern, (b) Raman spectrum and (c) XPS curve of active carbon cloth (ACC).



Fig. S3 (a) Ni 2p core-level spectrum and (b) O 1s core-level spectrum of sample Ni. (c) Co 2p core-level spectrum and (d) O 1s core-level spectrum of sample Co.



**Fig. S4** (a) CV curve of sample CoNi at 5 mV s<sup>-1</sup>. (b) Mass capacitance as a function of current density of different samples. Among various electrodes, CoNi electrode shows the best rate ability of ~ 68% compared to 18 % of Ni electrode and ~65 % of Co electrode.



Fig. S5 Thickness of the as-prepared electrodes.

1	1 1	
<b>Electrode materials</b>	Volumetric capacitance/ (F cm <sup>-3</sup> )	Reference
Mxene/CNT paper	435	1
Graphene/porous carbon	376	2
$T_3C_2Tx$ paper	442	3
MoS <sub>2</sub> nanosheet	700	4
NiCoO <sub>2</sub> /Ni(OH) <sub>2</sub> /Co(OH) <sub>2</sub>	700	This work

Table S1 Comparison of volumetric capacitance with reported literatures

Table S2 Resistances of different samples from EIS curves

	$R_{SR}/\Omega$	$R_{CT}/\Omega$	Angle at low frequency/°
Co sample	0.78	0.33	86
Ni sample	0.81	7.53	22
CoNi sample	1.02	0.32	66



Fig. S6 CV curves of (a) and (b) sample Co, (c) sample Ni and (d) sample CoNi.



**Fig.S7** The linear dependence of log current on log sweep rate (based on Equation (10)) for (a) sample Co, (b) sample Ni and (c) sample CoNi at -0.4 V during the correspondingly anodic CV processes.



**Fig. S6** (a-c) SEM images of carbon cloth with large surface area (CC). The corresponding (d) Raman spectrum, (e) XPS full pattern and (f) Nitrogen adsorption/desorption isotherm curve, inset: pore size distribution spectrum.



Fig. S7 (a) GCD curve, (b) rate ability and (c) Nyquist plots of CC as negative electrode.



Figure S8. Thickness of CC negative electrode.

Supercapacitor	Energy density/(mWh cm <sup>-3</sup> )	Reference
CoNi//CC using 6 M KOH	37.41	This work
CoNi//CC using KOH/PVA	11.98	This work
MnO <sub>2</sub> MN//Co <sub>3</sub> O <sub>4</sub>	27	5
MnO <sub>2</sub> /G//NF	0.27	6
$MnO_2//Fe_2O_3$	0.41	7
H-TiO2@MnO2//H-TiO2@C	0.3	8
MnO <sub>2</sub> /G//VOS@C	0.87	9
MnO <sub>2</sub> @ZnO//rGO	0.234	10
ACC//ACC	9.4	11

Table S3 Volumetric energy density vs the reported value in literatures

## Reference

- [1] M. Q. Zhao, C. E. Ren, Z. Ling, M. R. Lukatskaya, C. Zhang, K. L. Van Aken, M. W. Barsoum and Y. Gogotsi, *Advanced materials*, 2015, 27, 339-345.
- Y. Tao, X. Xie, W. Lv, D. M. Tang, D. Kong, Z. Huang, H. Nishihara, T. Ishii, B. Li, D. Golberg,
  F. Kang, T. Kyotani and Q. H. Yang, *Scientific reports*, 2013, 3, 2975.
- M. R. Lukatskaya, O. Mashtalir, C. E. Ren, Y. Dall'Agnese, P. Rozier, P. L. Taberna, M. Naguib,
  P. Simon, M. W. Barsoum and Y. Gogotsi, *Science*, 2013, 341, 1502-1505.
- [4] M. Acerce, D. Voiry and M. Chhowalla, *Nature nanotechnology*, 2015, **10**, 313-318.
- [5] J. S. Lee, D. H. Shin and J. Jang, *Energy Environ. Sci.*, 2015, **8**, 3030-3039.
- [6] T. Zhai, F. Wang, M. Yu, S. Xie, C. Liang, C. Li, F. Xiao, R. Tang, Q. Wu, X. Lu and Y. Tong, *Nanoscale*, 2013, 5, 6790-6796.
- [7] X. Lu, Y. Zeng, M. Yu, T. Zhai, C. Liang, S. Xie, M. S. Balogun and Y. Tong, Advanced materials, 2014, 26, 3148-3155.
- [8] X. Lu, M. Yu, G. Wang, T. Zhai, S. Xie, Y. Ling, Y. Tong and Y. Li, Advanced materials, 2013,

**25**, 267-272.

- [9] T. Zhai, X. Lu, Y. Ling, M. Yu, G. Wang, T. Liu, C. Liang, Y. Tong and Y. Li, Advanced materials, 2014, 26, 5869-5875.
- [10] W. Zilong, Z. Zhu, J. Qiu and S. Yang, J. Mater. Chem. C, 2014, 2, 1331-1336.
- [11] T. Qin, S. Peng, J. Hao, Y. Wen, Z. Wang, X. Wang, D. He, J. Zhang, J. Hou and G. Cao, Advanced Energy Materials, 2017, DOI: 10.1002/aenm.201700409, 1700409.