ADVANCED FUNCTIONAL MATERIALS

Supporting Information

for *Adv. Funct. Mater.*, DOI: 10.1002/adfm.201802016

Finely Crafted 3D Electrodes for Dendrite-Free and High-Performance Flexible Fiber-Shaped Zn–Co Batteries

Ming Li, Jiashen Meng, Qi Li, Meng Huang, Xiong Liu, Kwadwo Asare Owusu, Ziang Liu, and Liqiang Mai**

Supporting Information

Finely-Crafted 3D Electrodes for Dendrite-Free and High-Performance Flexible Fiber-Shaped Zn-Co Battery

Ming Li, Jiashen Meng, Qi Li, Meng Huang, Xiong Liu, Kwadwo Asare Owusu, Ziang Liu, Liqiang Mai**

Mr. M. Li, Mr. J. S. Meng, Prof. Q. Li, Mr. M. Huang, Mr. X. Liu, Mr. K. A. Owusu, Mr. Z. Liu, Prof. L. Q. Mai
State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Hubei, Wuhan 430070, P. R. China
E-mail: mlq518@whut.edu.cn; qi.li@whut.edu.cn

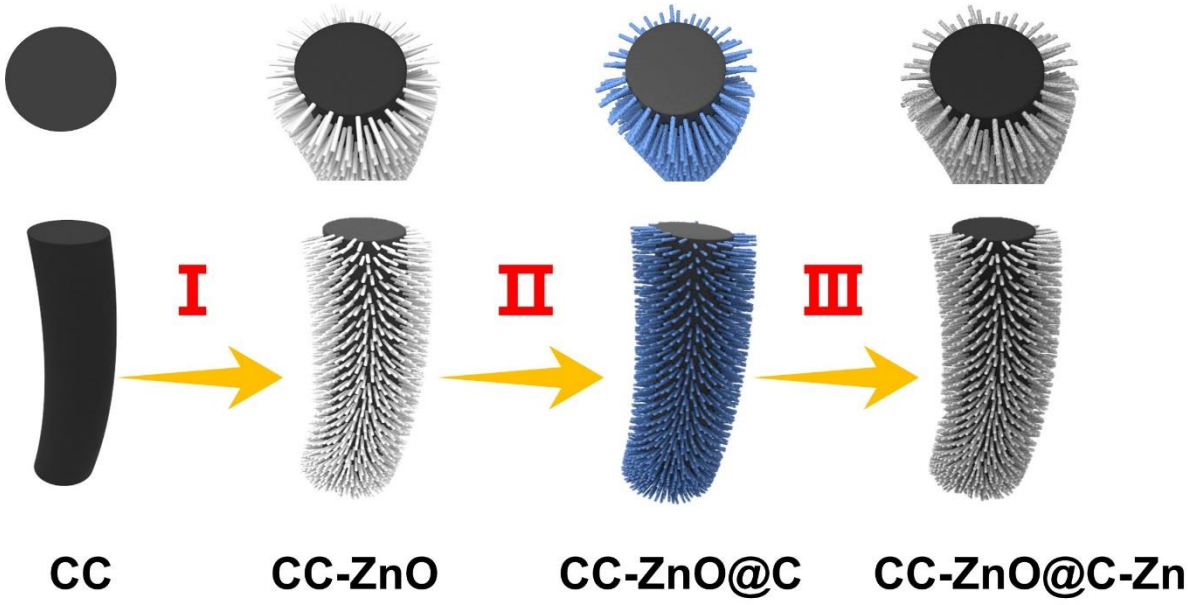

Figure S1. Schematic illustration of the synthesis process of CC-ZnO@C-Zn anode.

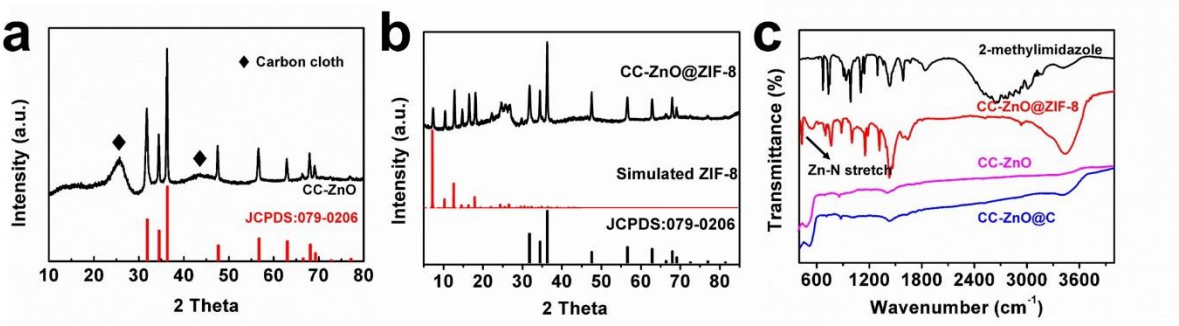

Figure S2. XRD patterns of a) CC-ZnO and b) CC-ZnO@ZIF. c) FT-IR spectra of 2-methylimidazole, CC-ZnO@ZIF-8, CC-ZnO and CC-ZnO@C.

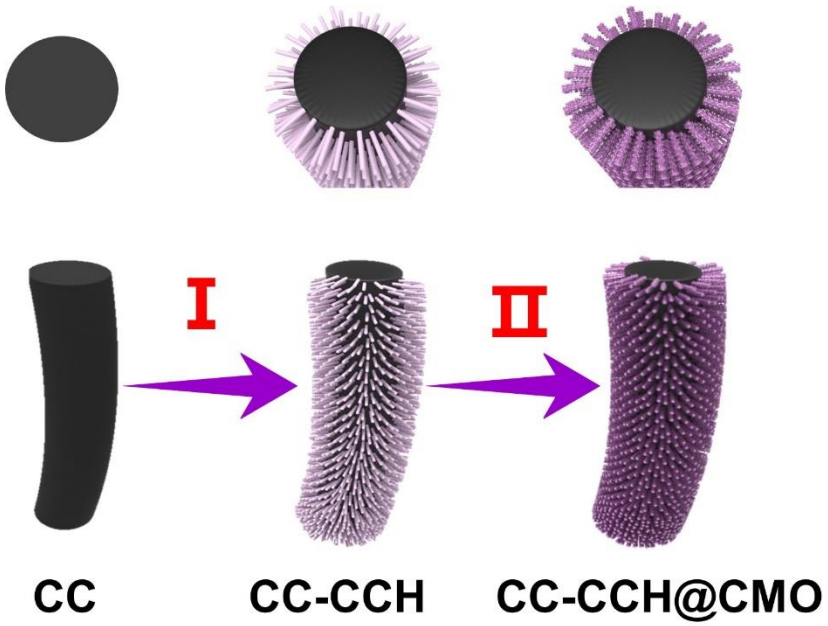

Figure S3. Schematic illustration of the synthesis process of the CC-CCH@CMO cathode.

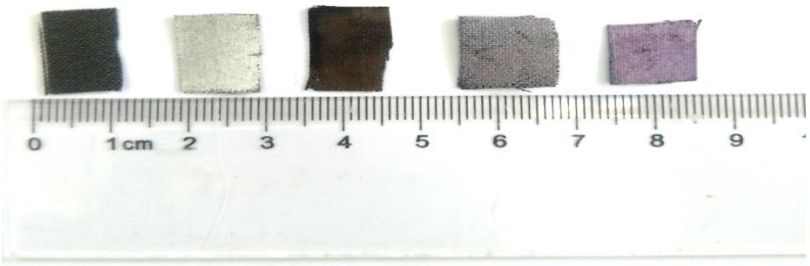

Figure S4. Optical photographs of as prepared samples. From left to right in turn are bare CC, CC-ZnO, CC-ZnO@C, CC-CCH and CC-CCH@CMO, respectively.

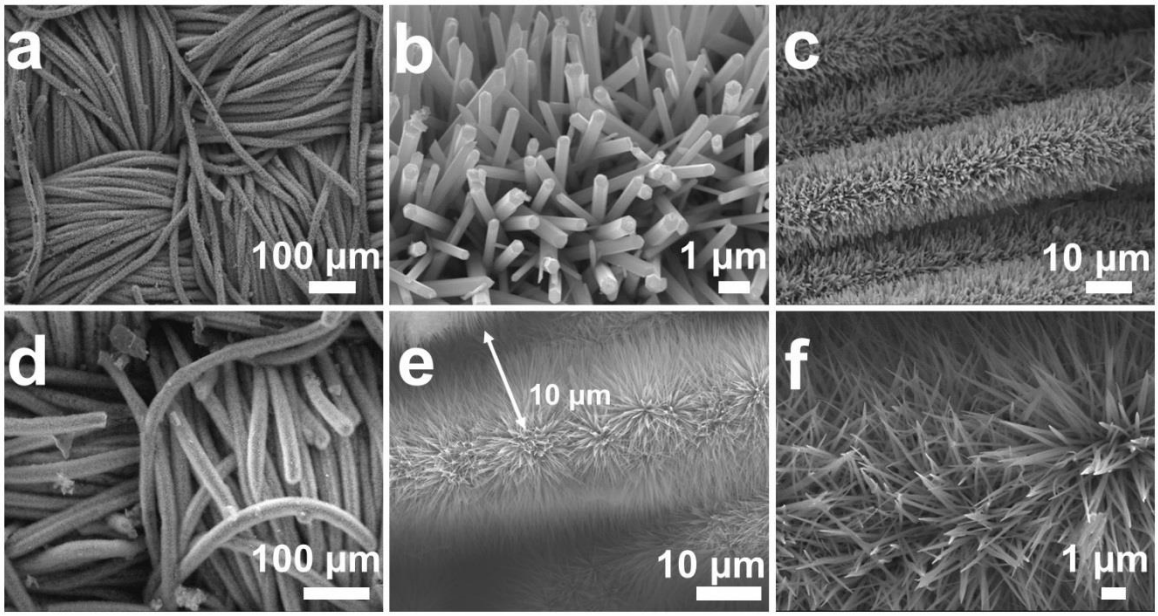

Figure S5. SEM images of a,b,c) CC-ZnO and d,e,f) CC-CCH.

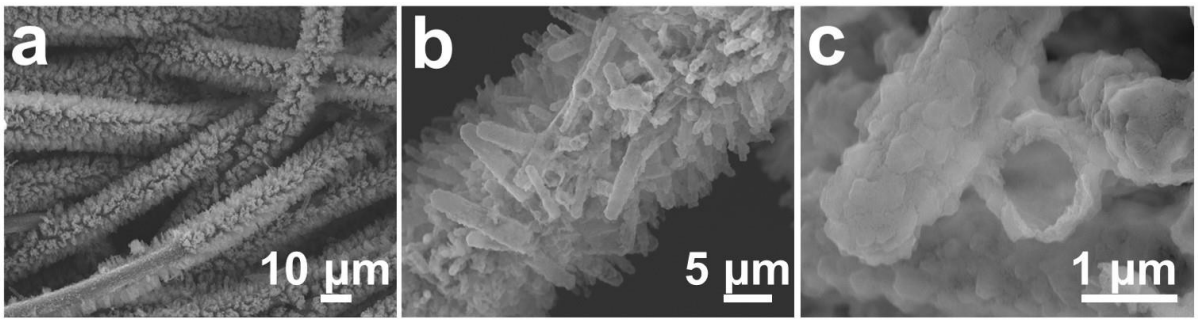

Figure S6. SEM images of ZIF-derived carbon tubes on CC.

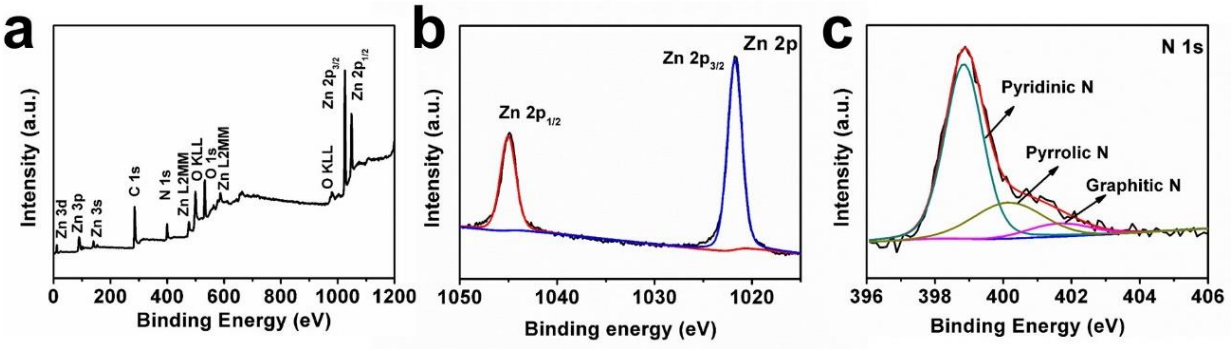

Figure S7. a) XPS full spectrum, b) Zn 2p spectrum and c) N 1s spectrum of CC-ZnO@C.

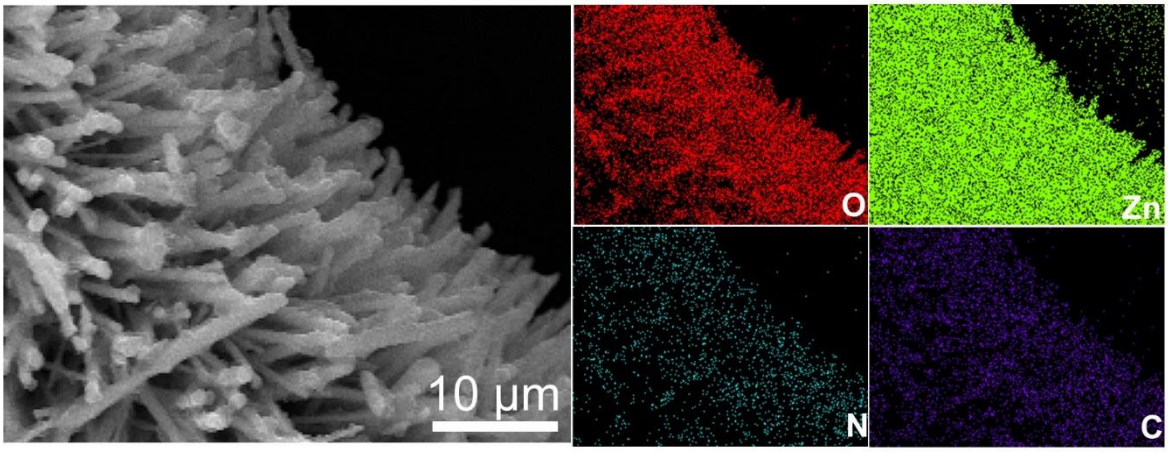

Figure S8. SEM image together with the corresponding elemental mapping images of CC-ZnO@C.

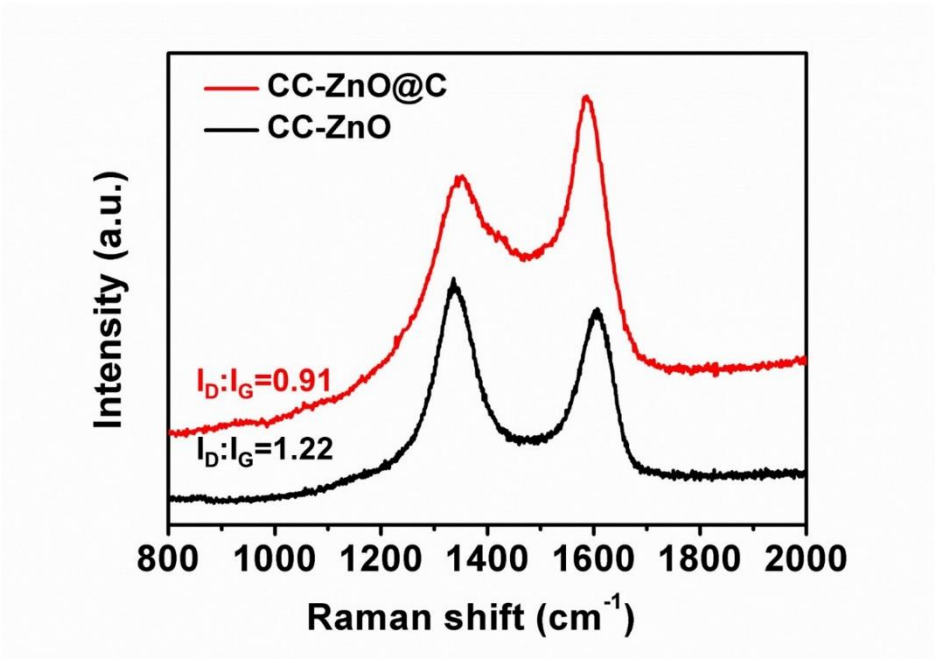

Figure S9. Raman plots of CC-ZnO@C and CC-ZnO.

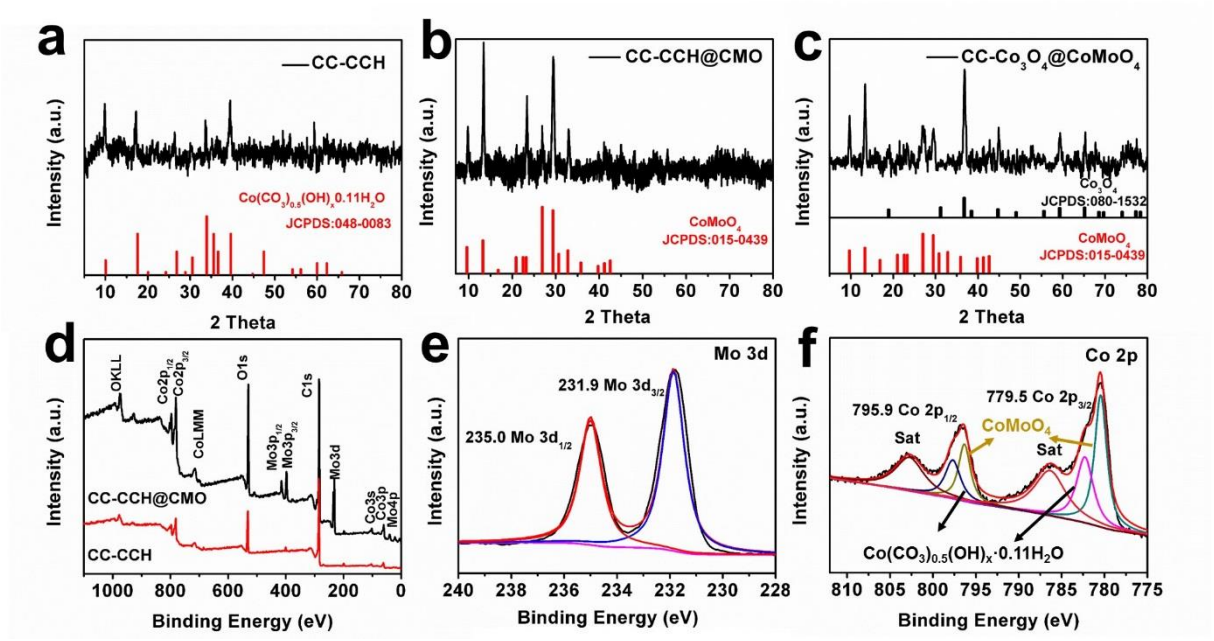

Figure S10. The XRD patterns of a) CC-CCH, b) CC-CCH@CMO and c) CC-Co_3O_4@$CoMoO_4$. d) XPS full spectra of CC-CCH@CMO and CC-CCH. e) Mo 3d spectrum, f) Co 2p spectrum of CC-CCH@CMO.

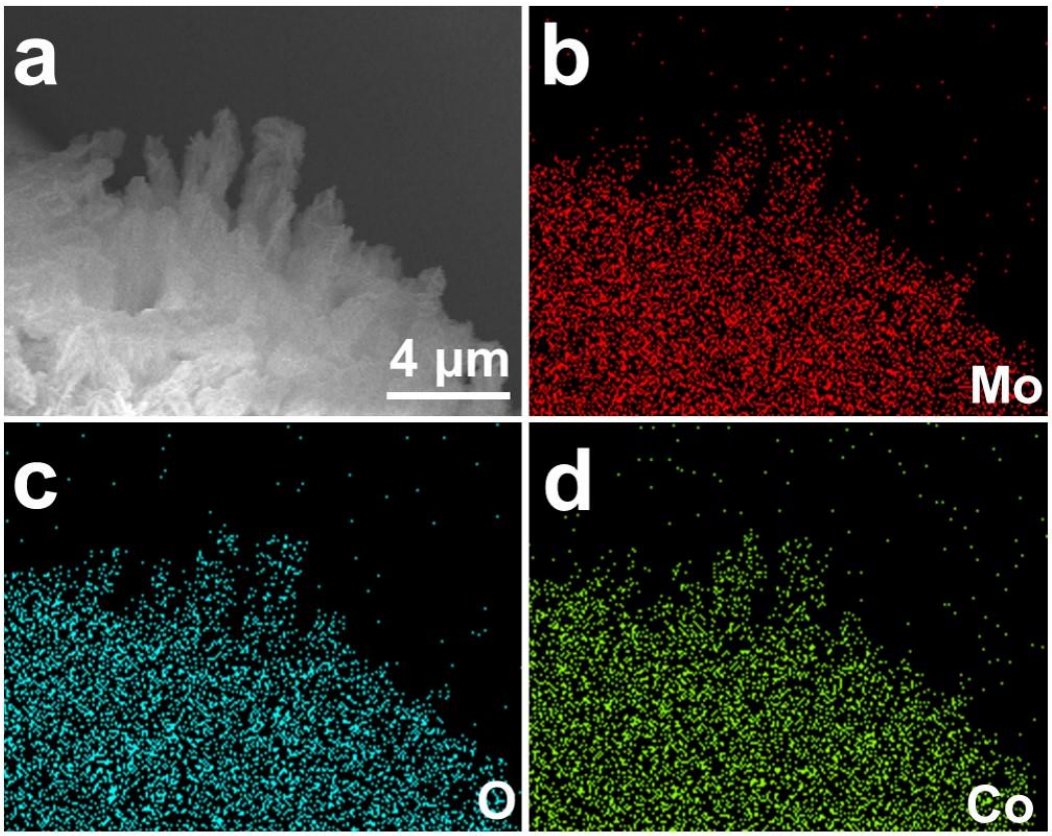

Figure S11. SEM image and the corresponding elemental mapping images of CC-CCH@CMO.

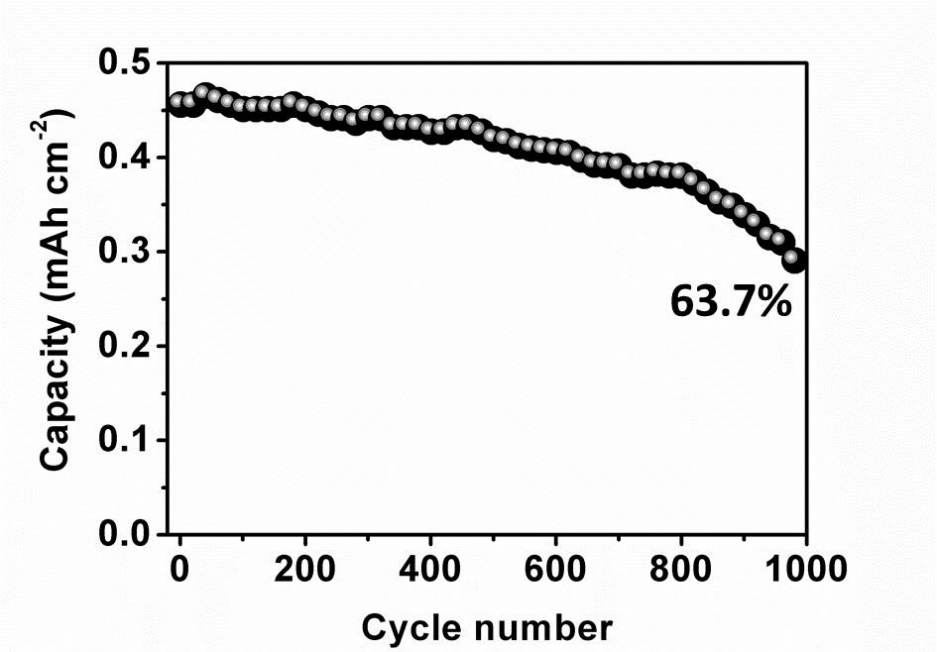

Figure S12. Cycling performance of aqueous Zn-Co full battery using the Zn plate as the anode at 40 mA cm^{-2}.

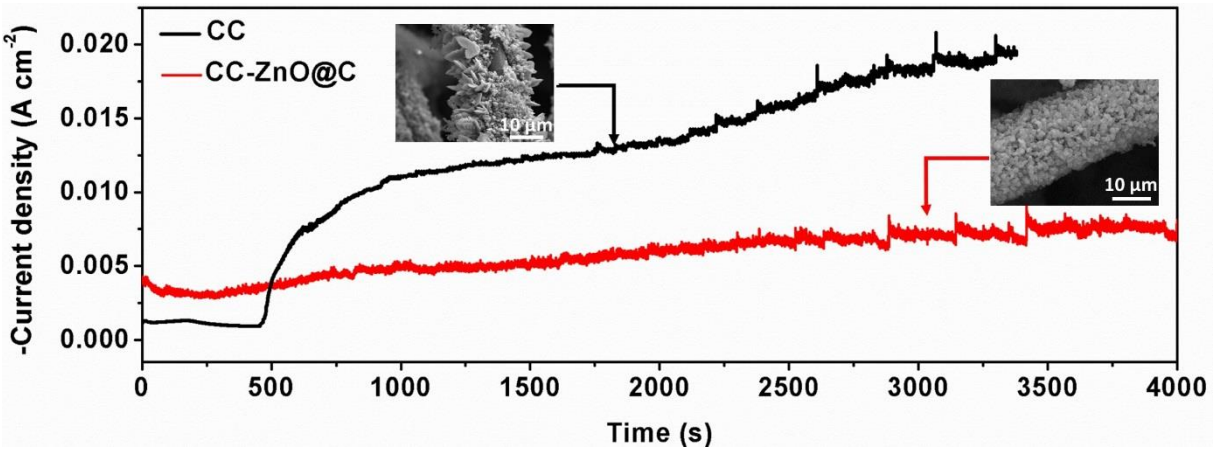

Figure S13. The current-time (i-t) curves of CC and CC-ZnO@C, and corresponding SEM images after Zn deposition.

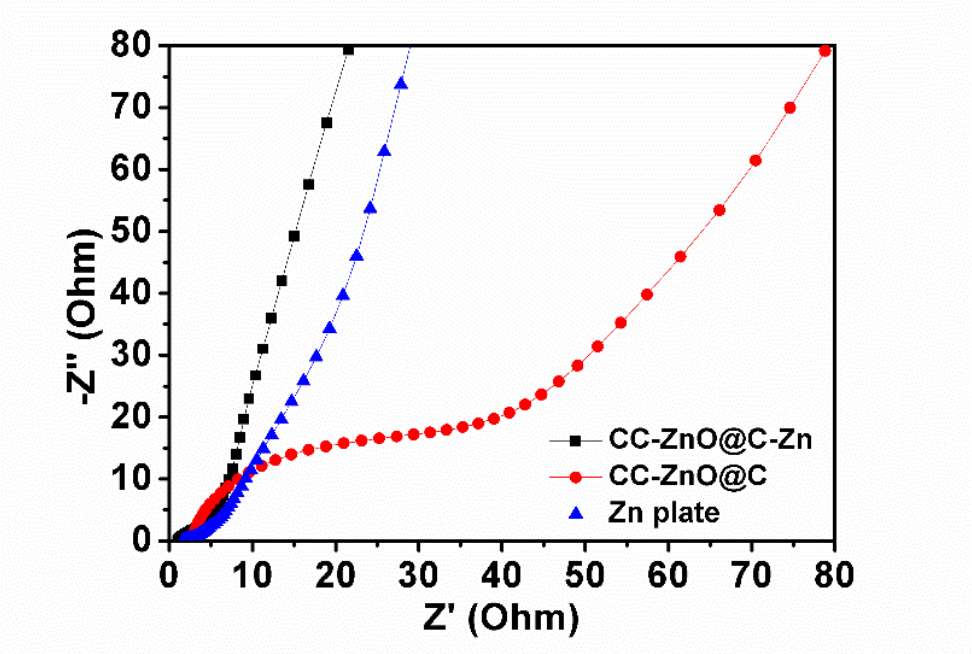

Figure S14. Comparison of the EIS results of CC-ZnO@C-Zn, CC-ZnO@C, and Zn plate.

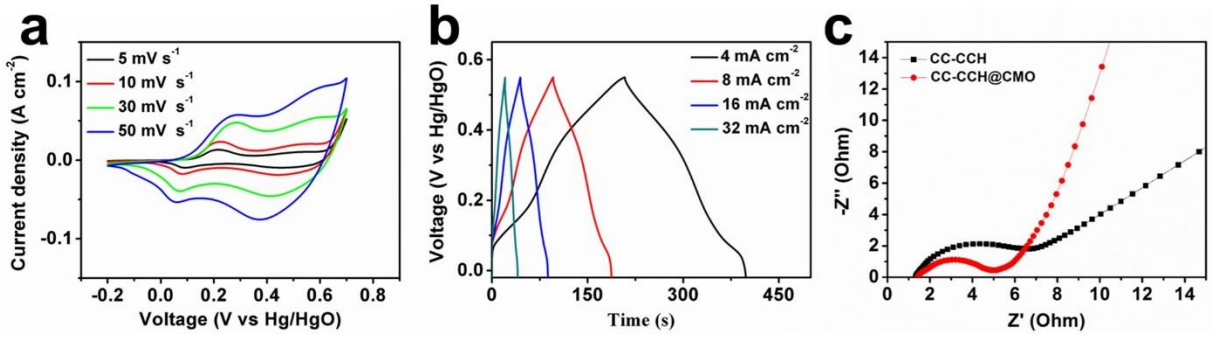

Figure S15. a) CV curves, and b) charge-discharge curves of CC-CCH. c) EIS results of CC-CCH@CMO and CC-CCH.

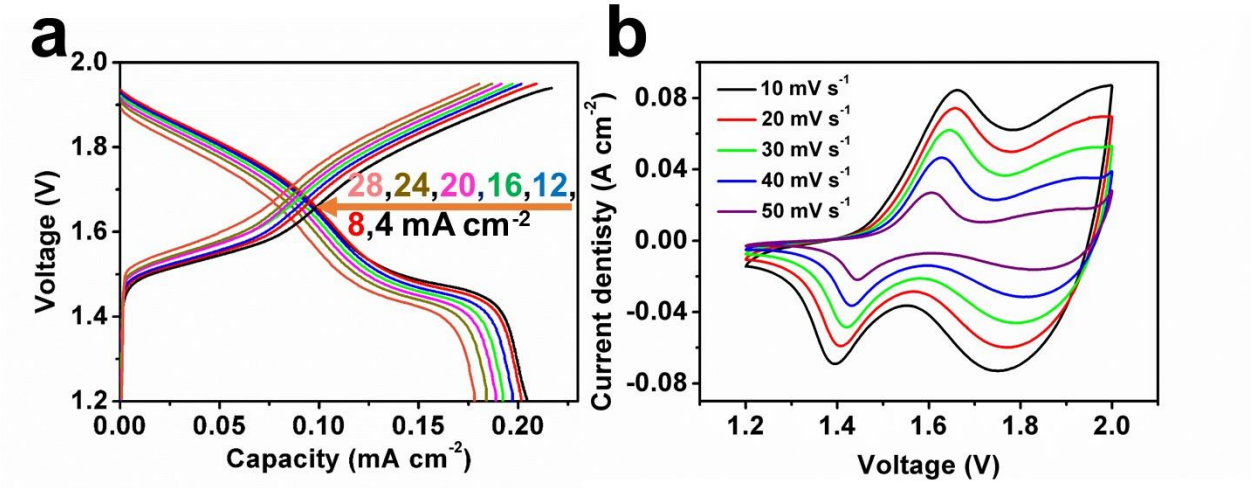

Figure S16. a) Charge-discharge curves, b) CV curves of aqueous Zn-Co full battery using the CC-CCH as the cathode**.**

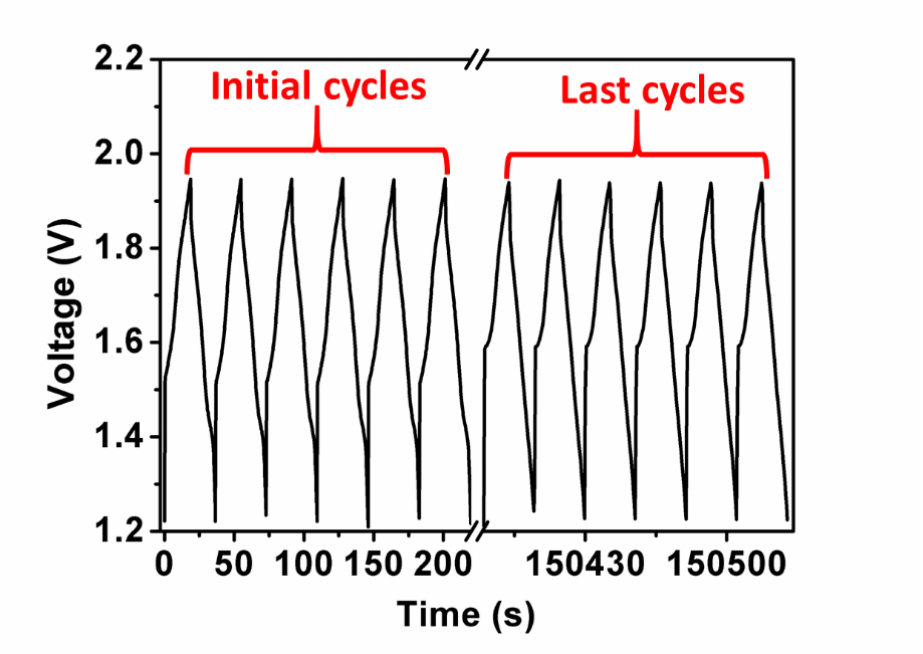

Figure S17. Charge-discharge curves of the initial and the last several cycles during the cycling test.

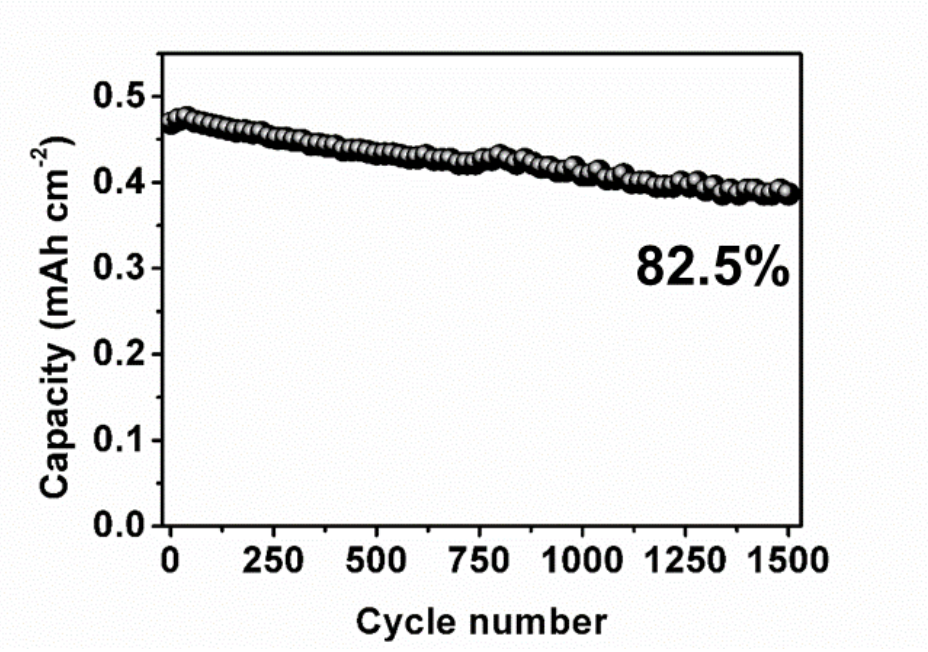

Figure S18. Cycling performance of the aqueous Zn-Co full battery using CC-ZnO@C-Zn as the anode and CC-CCH@CMO as the cathode at 40 mA cm^{-2}.

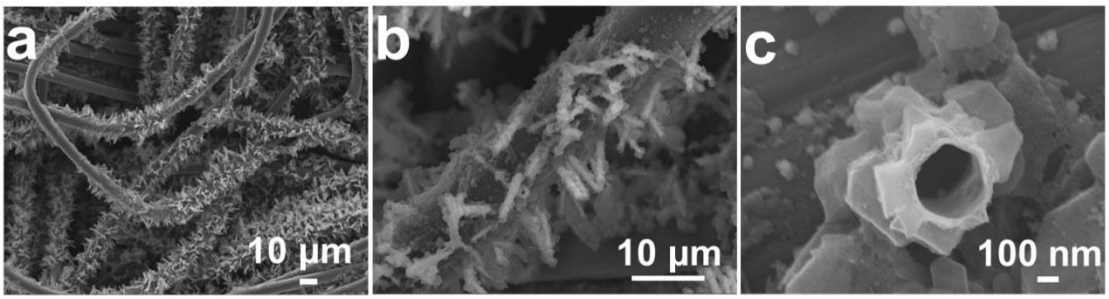

Figure S19. SEM images of 3D skeleton after 1500 cycles at 40 mA cm^{-2}.

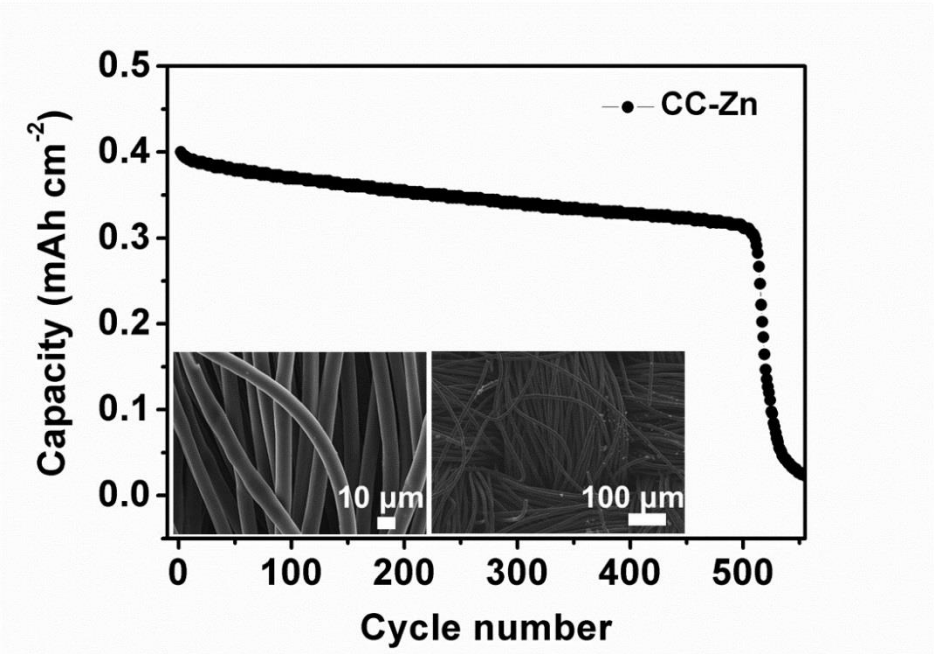

Figure S20. a) Cycling performance of aqueous Zn-Co battery using CC-Zn as anode at 40 mA cm^{-2}. The inset images are the SEM images of CC-Zn anode after cycling test.

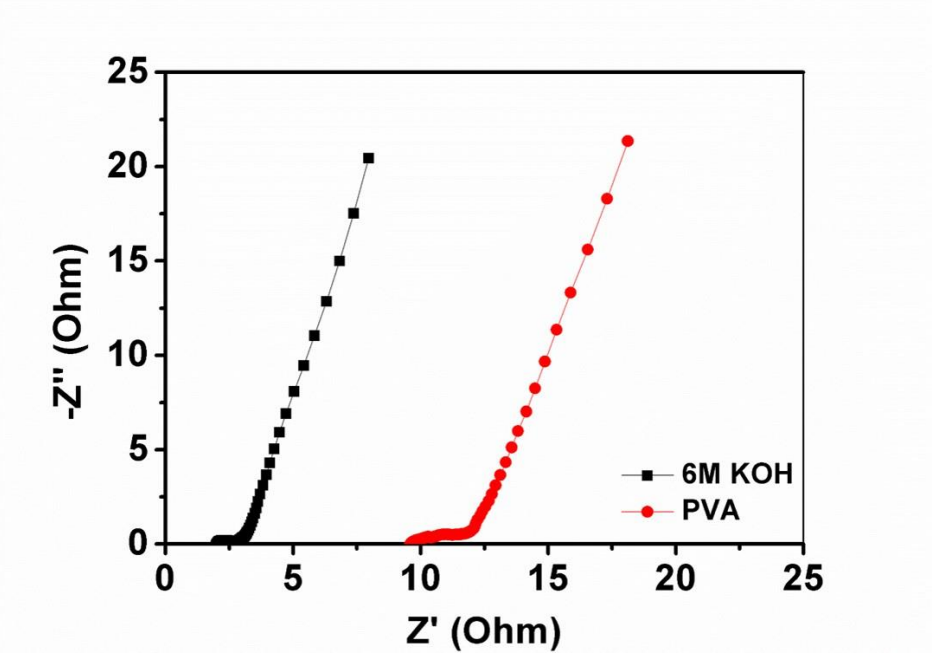

Figure S21. EIS results of Zn-Co battery using aqueous electrolyte (6 M KOH with 1.5 M ZnO) and solid-state (PVA-KOH) electrolyte.

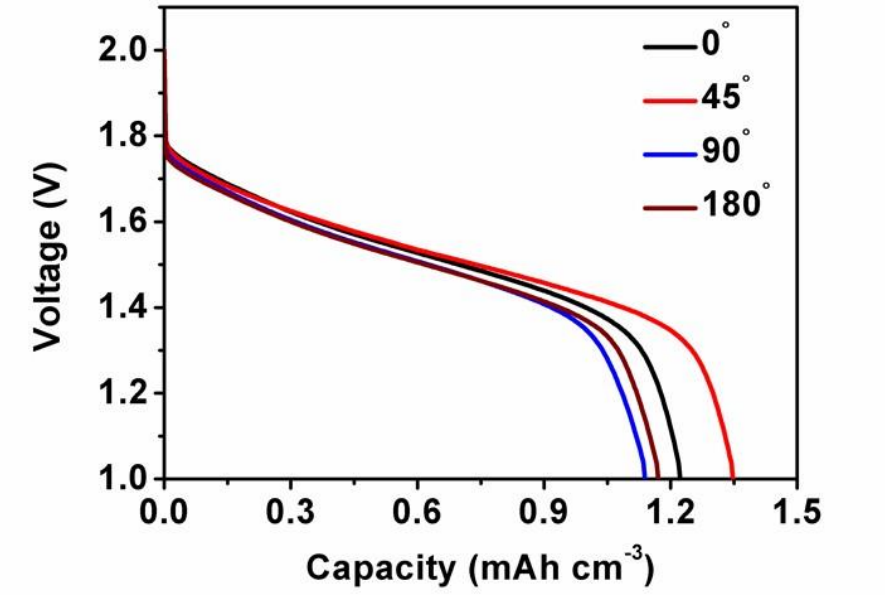

Figure S22. The capacity of fiber-shaped Zn-Co battery under various deformation states.

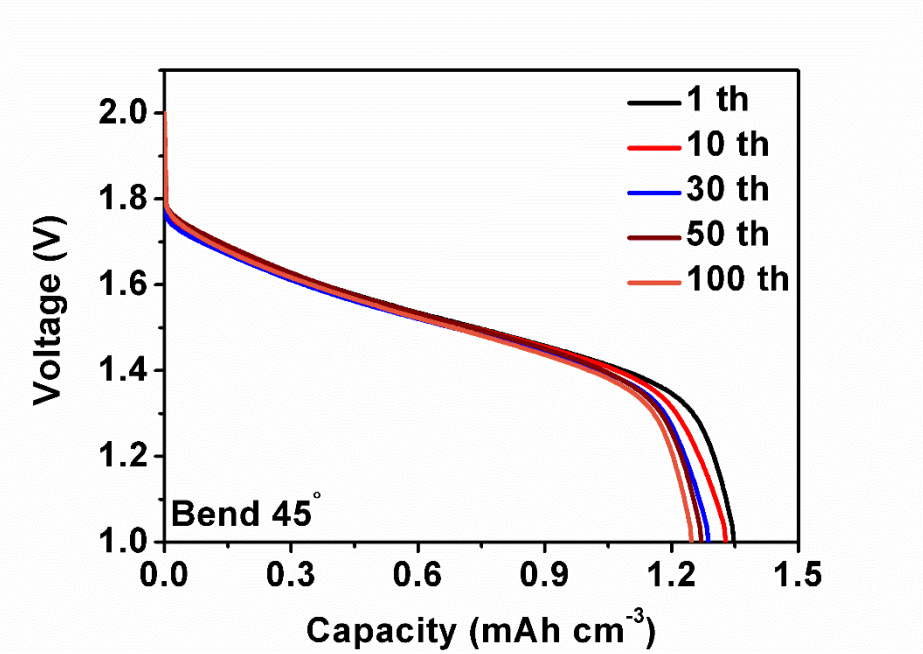

Figure S23. The capacity of fiber-shaped Zn-Co battery under a number of bending tests.

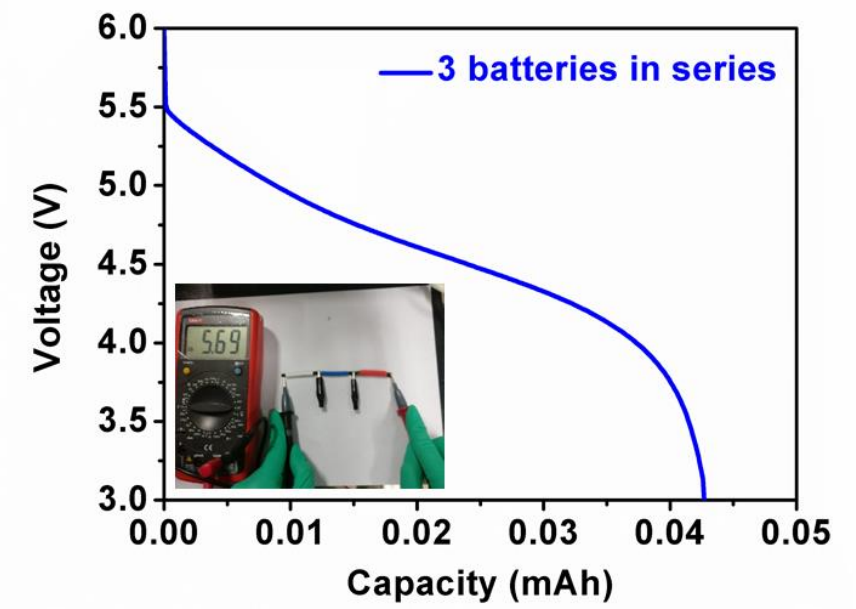

Figure S24. Galvanostatic discharge curves of a three-in-series battery. The inset digital image is the voltage output of the three-in-series batteries.

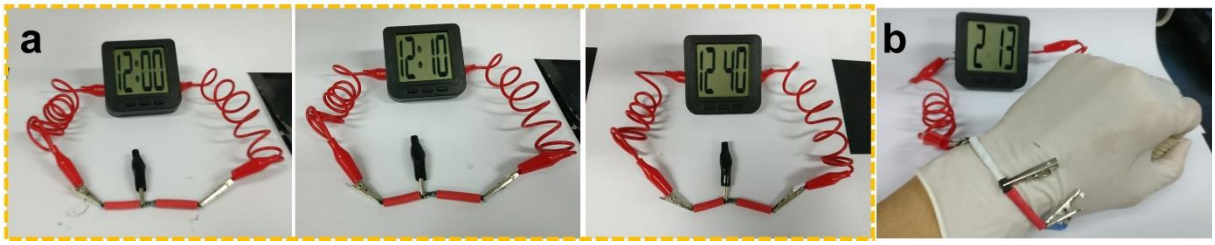

Figure S25. a) A digital watch was powered by two-in-series battery. b) A digital watch was powered by a bracelet battery.

Table S1. Comparison of our Zn-Co battery with other aqueous batteries reported before.

Aqueous Batteries	Active material Mass (mg)	Specific Capacity (mAh g^{-1})	Capacity Retention (%)	Power Density (kW kg^{-1})	Energy Density (Wh kg^{-1})
Ni-Zn battery (CC-CF@NiO//CC-CF@ZnO)[1]	1.92	203	72.9% (2400 cycles)	17.9	355.7
Ni-Zn battery (NiAlCo LDH/CNT//Zn) [2]	1.66	354	94% (2000 cycles)	16	274
Zn-Mn_2 battery[3]	5.38	366.6	83.7% (300 cycles)	8.6	504
Ni-Fe battery (CC-CF@NiO//CC-CF@Fe_3O_4)[4]	4.4	88.6	70.5 (2000 cycles)	1.2	94.5
Ni-based batteries (GF/CNTs/Fe_2O_3-$Ni(OH)_2$)[5]	1.5	119	89.1% (1000 cycles)	0.287	100.7
Ni-Bi battery (Ni−Co LDH// Bi_2O_3)[6]	2	110	93% (1000 cycles)	0.44	88.1
Ni-Zn battery (Zn–NiO)[7]	Not reported	155	65% (500 cycles)	Not reported	228
Zn-ion batteries[8]	Not reported	220	66.6% (35 cycles)	Not reported	Not reported
Na-Zn hybrid aqueous battery[9]	Not reported	76.2	81% (1000 cycles)	Not reported	62.9
Zn//VS_2 batteries[10]	Not reported	190.3	98% (200 cycles)	Not reported	123
Zn//Co_3O_4 Battery[11]	2	160	80% (2000 cycles)	Not reported	241
Rocking-Chair NH_4-Ion Battery[12]	Not reported	158.9	67% (1000 cycles)	Not reported	43
Aqueous Lithium-Ion Battery[13]	Not reported	40	82% (200 cycles)	Not reported	60
Ni-Bi battery ($NiCo_2O_4$//Bi)[14]	1.41	90	89% (1000 cycles)	21	85.8
Our work	4.98	143.2	71.1% (5000 cycles)	12.6 (aqueous) 10.5 (solid-state)	235 (aqueous) 221.9 (solid-state)

Table S2. Comparison of our flexible fiber-shaped Zn-Co battery with other reported before.

Flexible devices	Power Density ($mW\ cm^{-3}$)	Energy Density ($mWh\ cm^{-3}$)
Ni-Zn battery[15]	82.2	2.1
$MnO2//Fe_2O_3$[16]	0.31	0.31
Ni/GF/H-$CoMoO_4$//Ni/GF/H-Fe_2O_3 [17]	150	1.13
$CNTs//Fe_3O_4$-C[18]	29	1.2
H-ZnO@MnO_2 Symmetrical supercapacitor[19]	2.44	0.04
$TiN//Fe_2N$[20]	300	0.2
PEDOT paper[21]	52	1
Graphene//Co_3O_4[22]	1200	0.4
VOx//VN[23]	800	0.4
CoO@PPy//AC[24]	100	1.3
TiO_2@MnO_2//TiO_2@C[25]	210	0.5
MnO_2//WON[26]	600	1.1
MnO_2//Ti-Fe_2O_3@PEDOT[27]	520	0.6
Fiber-Shaped Ni–Zn Battery[28]	220	0.67
Our work	420	4.6

Reference

[1] J. Liu, C. Guan, C. Zhou, Z. Fan, Q. Ke, G. Zhang, C. Liu, J. Wang, *Adv. Mater.* **2016**, 28, 8732.

[2] M. Gong, Y. Li, H. Zhang, B. Zhang, W. Zhou, J. Feng, H. Wang, Y. Liang, Z. Fan, J. Liu, H. Dai, *Energy Environ. Sci.* **2014**, 7, 2025.

[3] Y. Zeng, X. Zhang, Y. Meng, M. Yu, J. Yi, Y. Wu, X. Lu, Y. Tong, *Adv. Mater.* **2017**, 29. 1700274.

[4] C. Guan, W. Zhao, Y. Hu, Q. Ke, X. Li, H. Zhang, J. Wang, *Adv. Energy Mater.***2016**, 6, 1601034.

[5] J. Liu, M. Chen, L. Zhang, J. Jiang, J. Yan, Y. Huang, J. Lin, H. J. Fan, Z. X. Shen,

Nano Lett. **2014**, 14, 7180.
[6] X. Li, C. Guan, Y. Hu, J. Wang, *ACS Appl. Mater. Interfaces* **2017**, 9, 26008.
[7] X. Wang, M. Li, Y. Wang, B. Chen, Y. Zhu, Y. Wu, *J. Mater. Chem. A* **2015**, 3, 8280.
[8] W. Kaveevivitchai, A. Manthiram, *J. Mater. Chem. A* **2016**, 4, 18737.
[9] K. Lu, B. Song, J. Zhang, H. Ma, *J. Power Sources* **2016**, 321, 257.
[10] P. He, M. Yan, G. Zhang, R. Sun, L. Chen, Q. An, L. Mai, *Adv. Energy Mater.* **2017**, 7, 1601920.
[11] X. Wang, F. Wang, L. Wang, M. Li, Y. Wang, B. Chen, Y. Zhu, L. Fu, L. Zha, L. Zhang, Y. Wu, W. Huang, *Adv. Mater.* **2016**, 28, 4904.
[12] D. X. Wu, Y. Qi, J. J. Hong, Z. Li, A. S. Hernandez, P. X. Ji, *Angew. Chem. Int. Ed.* **2017**, 129, 13206.
[13] J. Y. Luo, Y. Y. Xia, *Adv. Funct. Mater.* **2007**, 17, 3877.
[14] Y. Zeng, Z. Lin, Y. Meng, Y. Wang, M. Yu, X. Lu, Y. Tong, *Adv. Mater.* **2016**, 28, 9188.
[15] Z. Lu, X. Wu, X. Lei, Y. Li, X. Sun, *Inorg. Chem. Front*. **2015**, 2, 184.
[16] P. Yang, Y. Ding, Z. Lin, Z. Chen, Y. Li, P. Qiang, M. Ebrahimi, W. Mai, C. P. Wong, Z. L. Wang, *Nano Lett.* **2014**, 14, 731.
[17] K. Chi, Z. Zhang, Q. Lv, C. Xie, J. Xiao, F. Xiao, S. Wang, *ACS Appl. Mater. Interfaces.* **2017**, 9, 6044.
[18] R. Li, Y. Wang, C. Zhou, C. Wang, X. Ba, Y. Li, X. Huang, J. Liu, *Adv. Funct. Mater.* **2015**, 25, 5384.
[19] P. Yang, X. Xiao, Y. Li, Y. Ding, P. Qiang, X. Tan, W. Mai, Z. Lin, W. Wu, T. Li, *ACS nano* **2013**, 7, 2617.
[20] C. Zhu, P. Yang, D. Chao, X. Wang, X. Zhang, S. Chen, B. K. Tay, H. Huang, H. Zhang, W. Mai, H. J. Fan, *Adv. Mater.* **2015**, 27, 4566.
[21] B. Anothumakkool, S. N. Bhange, R. Soni, S. Kurungot, *Energy Environ. Sci.* **2015**, 8, 1339.
[22] X. Wang, B. Liu, R. Liu, Q. Wang, X. Hou, D. Chen, R. Wang, G. Shen, *Angew. Chem.* Int. Ed **2014**, 53, 1849.
[23] X. Lu, M. Yu, T. Zhai, G. Wang, S. Xie, T. Liu, C. Liang, Y. Tong, Y. Li, *Nano Lett.* **2013**, 13, 2628.
[24] C. Zhou, Y. Zhang, Y. Li, J. Liu, *Nano Lett.* **2013**, 13, 2078.
[25] X. Lu, M. Yu, G. Wang, T. Zhai, S. Xie, Y. Ling, Y. Tong, Y. Li, *Adv. Mater.* **2013**, 25, 267.
[26] M. Yu, Y. Han, X. Cheng, L. Hu, Y. Zeng, M. Chen, F. Cheng, X. Lu, Y. Tong, *Adv. Mater.* **2015**, 27, 3085.
[27] Y. Zeng, Y. Han, Y. Zhao, Y. Zeng, M. Yu, Y. Liu, H. Tang, Y. Tong, X. Lu, *Adv. Energy. Mater.* **2015**, 5, 1402176.
[28] Y. Zeng, Y. Meng, Z. Lai, X. Zhang, M. Yu, P. Fang, M. Wu, Y. Tong, X. Lu, *Adv. Mater.* **2017**, 29, 1702698.