

## Supported Information

**Configuring hierarchical Ni/NiO 3D-network assisted with bamboo cellulose nanofibers  
for high-performance Ni-Zn aqueous batteries**

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### Calculations:

Specific capacity values of single electrode and battery were calculated from the galvanostatic discharge curves by the following equation:

$$C = \frac{It}{m}$$

where C is specific capacity (mAh.g<sup>-1</sup>), m refers to the loading mass of active materials (g), I corresponds to the discharging density (A), t represents the discharge time (h).

The energy density (E) and power density (P) were calculated from the equations as below:

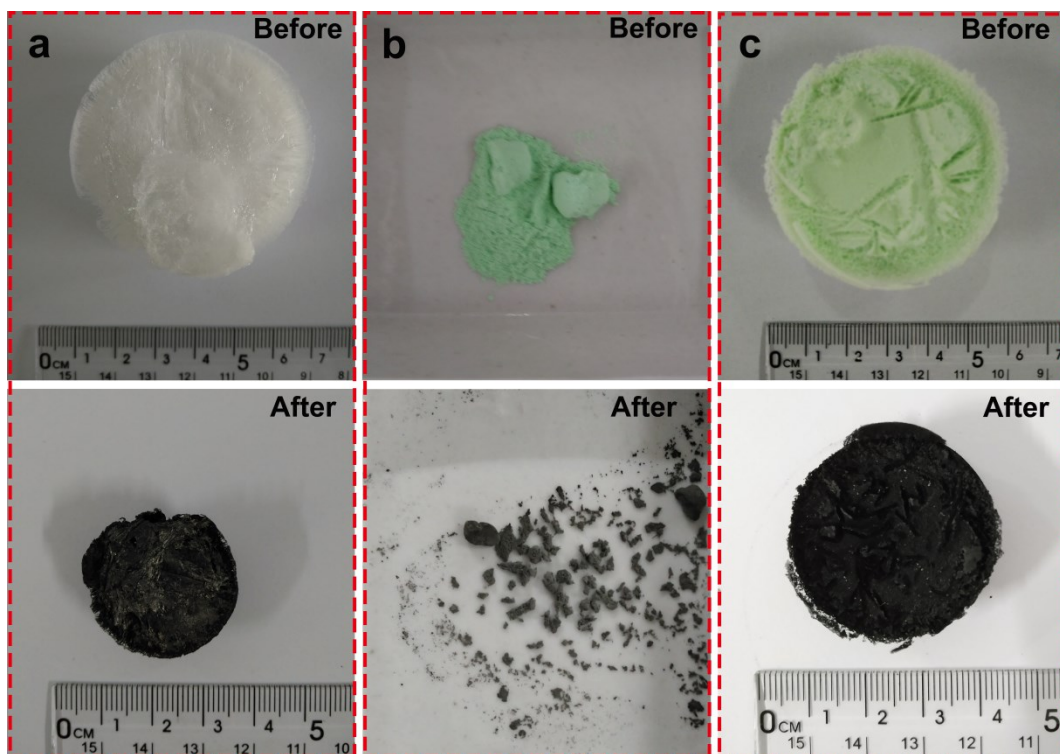
$$E = \frac{\int_0^{\Delta t} IV(t) dt}{\Delta t}$$
$$P = \frac{E}{1000 \times \Delta t}$$

where the unit of E is Wh.kg<sup>-1</sup> and that of P is kW.kg<sup>-1</sup>.  $\Delta t$  is the discharge time (h), I corresponds to the discharging density (A), V refers to the discharging voltage (V) and dt represents time differential.

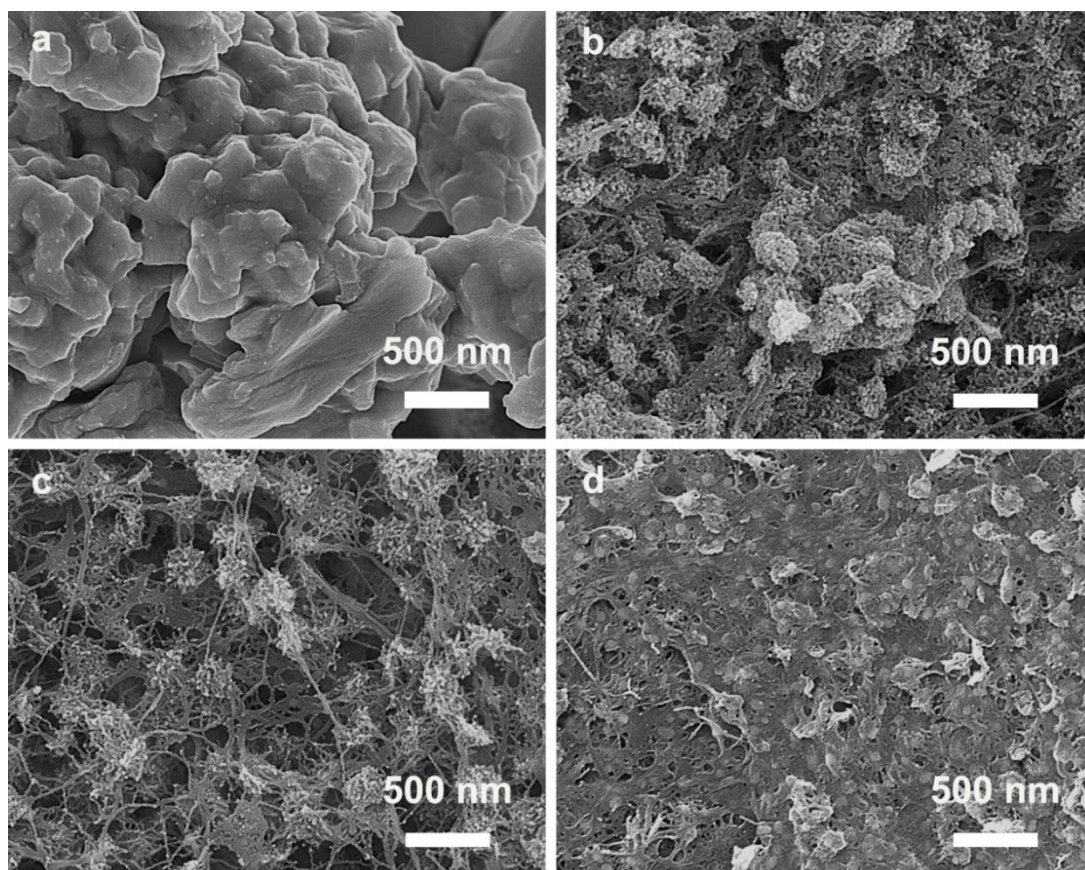
The mass ratios of bamboo cellulose nanofibers to the nickel of Ni(NO<sub>3</sub>)<sub>2</sub> were calculated from the following equations:

$$m_r = \frac{m_1}{n \times N \div 1000}$$

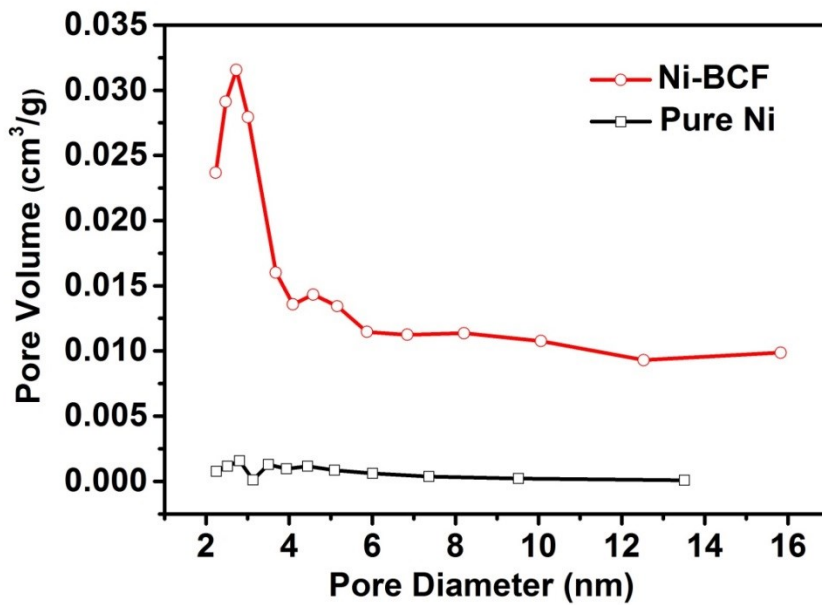
where the unit of  $m_r$  is g/g.  $m_1$  is the mass of bamboo cellulose nanofibers (g), which is 0.2 in this experiment.  $n$  is the amount of Ni(NO<sub>3</sub>)<sub>2</sub> (mmol, and  $n=2,4,6$ ).  $N$  is the molar mass of nickel ( $N=58.69$ ).



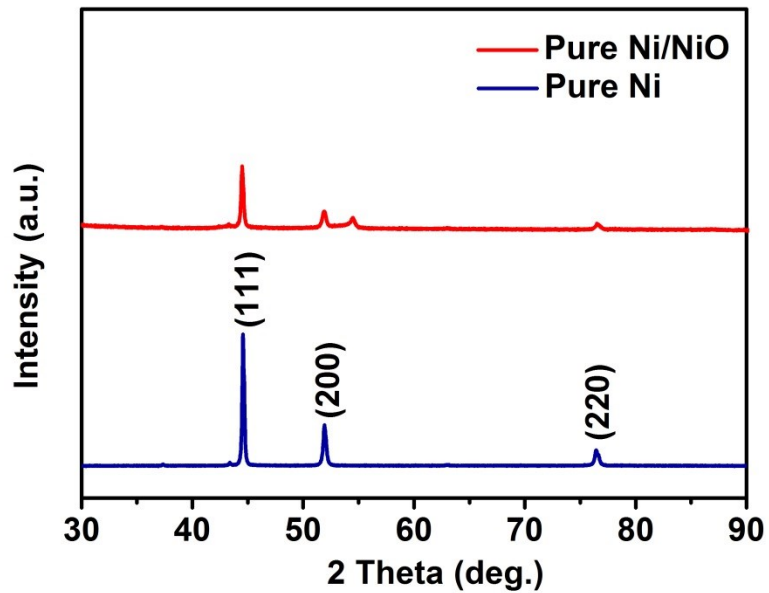
**Figure S1.** The macroscopic morphology of (a) BCF, (b) pure nickel, and (c) Ni-BCF before and after calcination



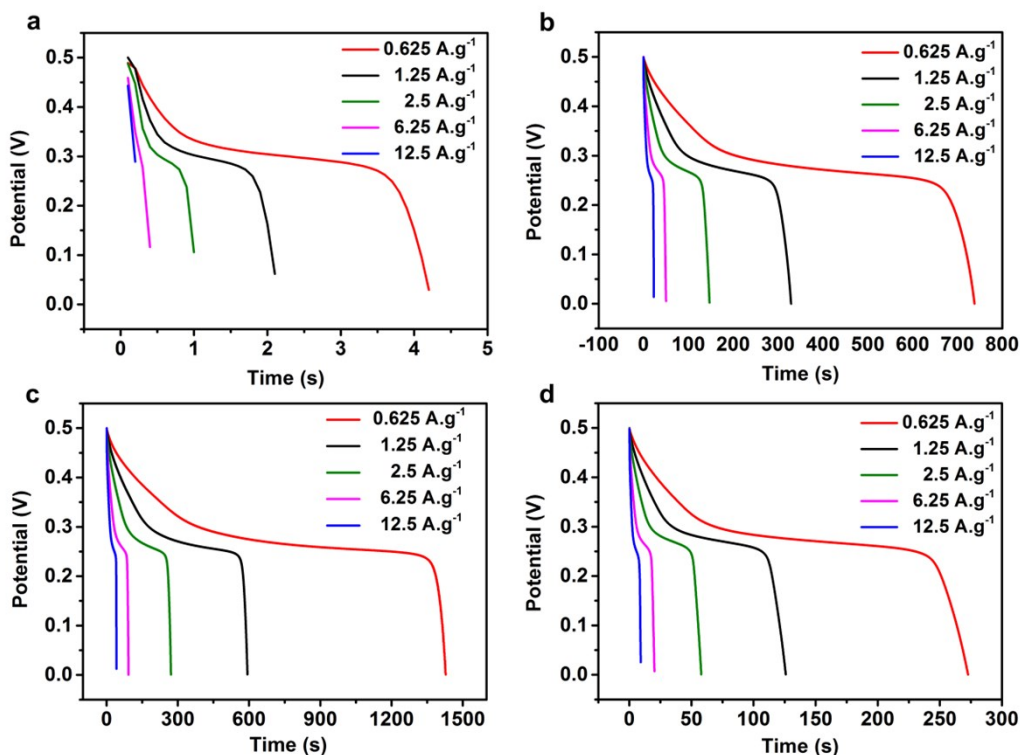
**Figure S2.** The SEM images of (a) pure Ni, (b) Ni-BCF0.57, (c) Ni-BCF0.85 and (d) Ni-BCF1.7



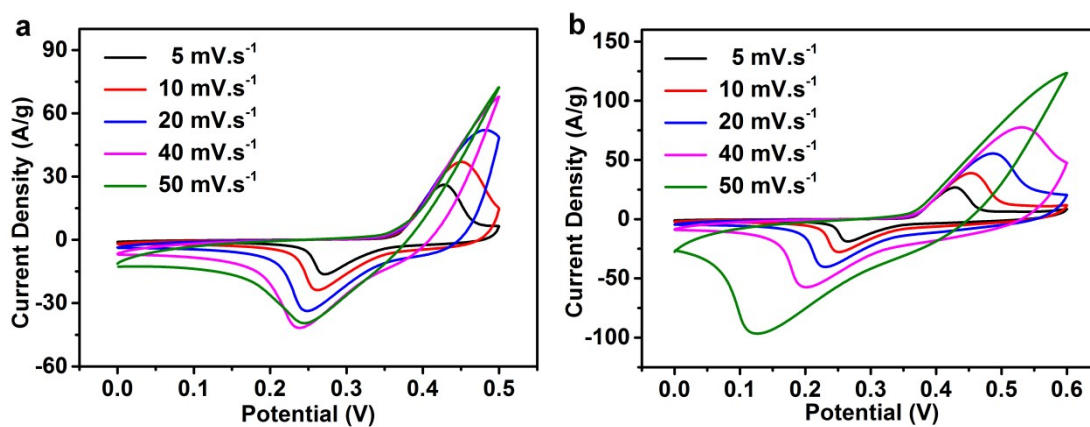
**Figure S3.** The pore size distribution of Ni-BCF and pure Ni



**Figure S4.** XRD pattern of pure Ni and pure Ni/NiO



**Figure S5.** The discharge curves of (a) pure Ni, (b) Ni-BCF<sub>0.57</sub>, (c) Ni-BCF<sub>0.85</sub>, and (d) Ni-BCF<sub>1.7</sub> at different current densities



**Figure S6.** CV curves of Ni/NiO-BCF<sub>0.85</sub> electrode with different potential window of (a) 0-0.5V and (b) 0-0.6V

**Table S1.** Cycling performance of different nickel-based electrodes

Electrode structure	Electrolyte	Cycling stability	Reference
Ni/NiO-BCF hierarchical networks	6 M KOH	94.5% after 2000 cycles	<b>This work</b>
Porous net-like $\beta$ -Ni(OH) <sub>2</sub> / $\gamma$ -NiOOH	1 M KOH	81% after 1000 cycles	1

NiCo <sub>2</sub> O <sub>4</sub> @MnO <sub>2</sub> core-shell nanowire array	1 M LiOH	88% after 2000 cycles	2
Ni-MOF nanosheet/carbon nanotube fiber	2 M KOH	79.6% after 2000 cycles	3
Co <sub>3</sub> O <sub>4</sub> @NiO nanostrip@nanorod arrays	6 M KOH	96% after 1000 cycles	4
Ni <sub>3</sub> S <sub>2</sub> honeycomb-like nanosheet	1 M KOH	89% after 1000 cycles	5
Ni(OH) <sub>2</sub> nanosheets/nitrogen-doped graphene	1 M KOH	93% after 1000 cycles	6

### References:

- [1] Yuan Y, Xia X, Wu J, Yang J, Chen Y, Guo SY. Nickel foam-supported porous Ni(OH)<sub>2</sub>/NiOOH composite film as advanced pseudocapacitor material. *Electrochimica Acta*, 2011, 56(6): 2627-2632.
- [2] Yu L, Zhang G, Yuan C, Lou X. Hierarchical NiCo<sub>2</sub>O<sub>4</sub>@MnO<sub>2</sub> core-shell heterostructured nanowire arrays on Ni foam as high-performance supercapacitor electrodes. *Chemical Communication*, 2013, 49(2): 137-139.
- [3] Li C, Zhang Q, Li T, He B, Man P, Zhu Z, Zhou Z, Wei L, Zhang K, Hong G, Yao Y. Nickel metal-organic framework nanosheets as novel binder-free cathode for advanced fibrous aqueous rechargeable Ni-Zn battery. *Journal of Materials Chemistry A*, 2020, 8(6): 3262-3269.
- [4] Lu Z, Wu X, Lei X, Li Y, Sun X. Hierarchical nanoarray materials for advanced nickel-zinc batteries. *Inorganic Chemistry Frontiers*, 2015, 2(2): 184-187.
- [5] Fu W, Zhao Y, Mei J, Wang F, Han W, Wang F, Xie E. Honeycomb-like Ni<sub>3</sub>S<sub>2</sub> nanosheet arrays for high-performance hybrid supercapacitors. *Electrochimica Acta*, 2018, 283: 737-743.
- [6] Wu X, Wu HB, Xiong W, Le Z, Sun F, Liu F, Chen J, Zhu Z, Lu Y. Robust iron nanoparticles with graphitic shells for high-performance Ni-Fe battery. *Nano Energy*, 2016, 30: 217-224.

**Table S2.** Cycling performance of different aqueous rechargeable batteries

Battery	Electrolyte	Cycling stability	Reference
Ni/NiO//Zn	6 M KOH+0.5 M Zn(Ac) <sub>2</sub>	94.5% after 1000 cycles	<b>This work</b>

Ni-MOF/CNTF//Zn	2M KOH saturated with ZnO	89% after 600 cycles	1
Ni(OH) <sub>2</sub> //ZnO/C	6M KOH saturated with ZnO	89% after 800 cycles	2
NiCo <sub>2</sub> O <sub>4</sub> //Zn	6 M KOH+0.1M Zn(Ac) <sub>2</sub>	63.23% after 1000 cycles	3
NiO-CNTs//Zn	1 M KOH +10 mM Zn(Ac) <sub>2</sub>	65% after 500 cycles	4
Co <sub>3</sub> O <sub>4</sub> @NiO NSRAs//Zn	6M KOH saturated with ZnO	89% after 500 cycles	5
CC-CF@NiO//CC-CF@ZnO	2M KOH saturated with ZnO	91.45% after 1000 cycles	6
NCHO//Zn	6 M KOH	60% after 1000 cycles	7
Ni(OH) <sub>2</sub> /NG//Fe	1 M KOH	93% after 1000 cycles	8

### References:

- [1] Li C, Zhang Q, Li T, He B, Man P, Zhu Z, Zhou Z, Wei L, Zhang K, Hong G, Yao Y. Nickel metal–organic framework nanosheets as novel binder-free cathode for advanced fibrous aqueous rechargeable Ni–Zn battery. *Journal of Materials Chemistry A*, 2020, 8(6): 3262-3269.
- [2] Zeng X, Yang Z, Meng J, Chen L, Chen H, Qin H. The cube-like porous ZnO/C composites derived from metal organic framework-5 as anodic material with high electrochemical performance for Ni–Zn rechargeable battery. *Journal of Power Sources*, 2019, 438: 226986.
- [3] Lu Z, Wu X, Lei X, Li Y, Sun X. Hierarchical nanoarray materials for advanced nickel–zinc batteries. *Inorganic Chemistry Frontiers*, 2015, 2(2): 184-187.
- [4] Wang X, Li M, Wang Y, Chen B, Zhu Y, Wu Y. A Zn–NiO rechargeable battery with long lifespan and high energy density. *Journal of Materials Chemistry A*, 2015, 3(16): 8280-8283.
- [5] Huang M, Zhou X, Hou X, Wang S, Zhuang Y, Jia H, Guan M. Intermediate phase  $\alpha$ - $\beta$ -Ni<sub>1-x</sub>Co<sub>x</sub>(OH)<sub>2</sub>/carbon nanofiber hybrid material for high performance nickel-zinc battery. *Electrochimica Acta*, 2018, 298(1): 127-133.
- [6] Liu J, Guan C, Zhou C, Fan Z, Ke Q, Zhang G, Liu C, Wang J. A flexible quasi-solid-state nickel-zinc battery with high energy and power densities based on 3D electrode design. *Advance Materials*, 2016, 28(39): 8732-8739.

- [7] Huang Y, Ip WS, Lau YY, Sun J, Zeng J, Yeung NSS, Ng WS, Li H, Pei Z, Xue Q, Wang Y, Yu J, Hu H, Zhi C. Weavable, conductive yarn-based NiCo//Zn textile battery with high energy density and rate capability. *ACS Nano*, 2017, 11(9): 8953-8961.
- [8] Wu X, Wu H Xiong W, Le Z, Sun F, Liu F, Chen J, Zhu Z, Lu Y. Robust iron nanoparticles with graphitic shells for high-performance Ni-Fe battery. *Nano Energy*, 2016, 30: 217-224.