Supporting Information

Realizing high-rate aqueous zinc-ion battery using organic cathode material containing electron withdrawing group

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1. **Supporting Figures and Tables**

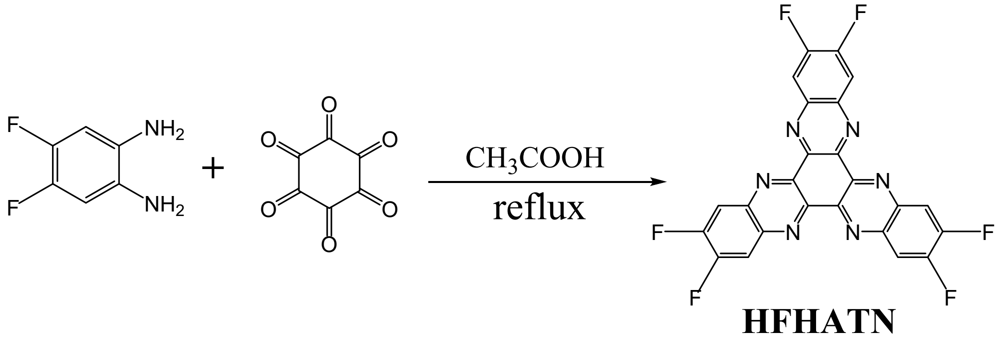


Figure S1. Synthetic route of HFHATN.

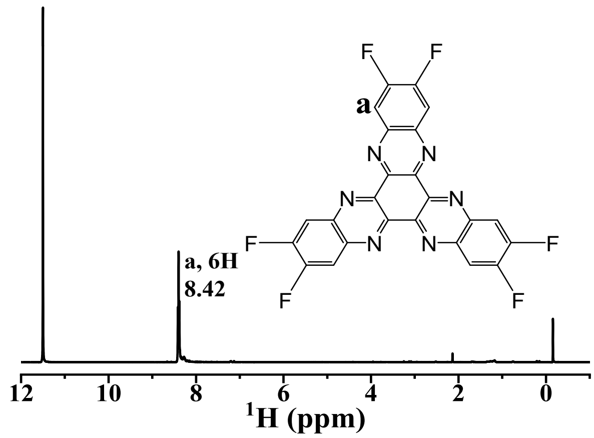


Figure S2. Liquid state ^{1}H NMR spectrum of HFHATN.

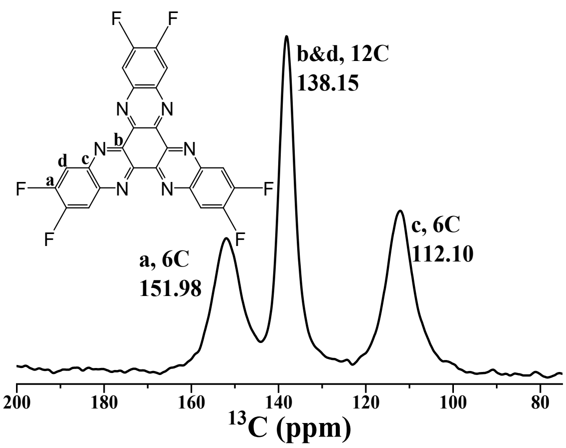
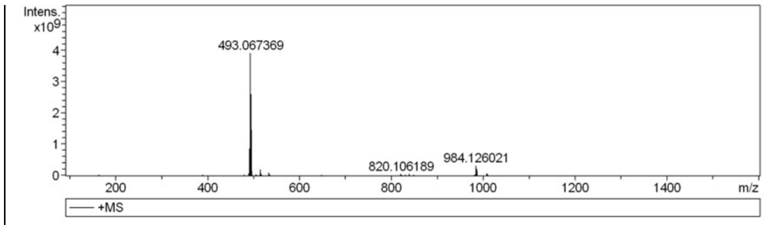


Figure S3. Solid state ^{13}C NMR spectrum of HFHATN.



#	m/z	I	FWHM	S/N	Res.
1	489.076695	62422712	0.002108	1766.6	232043
2	490.064567	75693808	0.002141	2142.5	228883
3	491.072061	293011232	0.002205	8299.0	222692
4	492.060400	870645568	0.002170	24663.2	226806
5	492.062828	59899736	0.001224	1695.1	402033
6	492.074190	58998616	0.002104	1669.6	233896
7	492.077217	93945688	0.002099	2659.6	234433
8	493.062333	56164240	0.001402	1589.3	351568
9	493.063624	104136592	0.001398	2948.3	352794
10	493.064869	235470736	0.000961	6668.9	513326
11	493.067369	3913351168	0.002170	110861.9	227202
12	493.072296	65994640	0.001695	1867.7	290813
13	494.068961	51576772	0.001783	1459.3	277102
14	494.070254	65994692	0.001333	1867.7	370609
15	494.071635	187236288	0.001367	5302.5	361308
16	494.073982	2600534016	0.002190	73670.3	225636
17	494.077724	71565256	0.001395	2025.6	354064
18	495.078370	574947328	0.002117	16286.2	233911
19	515.050848	203359248	0.002362	5645.8	218093
20	516.056691	63996988	0.002093	1775.5	246601
21	533.084281	112265040	0.002488	3115.9	214234
22	534.091200	63637368	0.002430	1765.5	219787
23	820.106189	61616736	0.005734	1367.0	143030
24	838.098388	54965564	0.006017	1219.3	139284
25	983.123168	111562776	0.008565	2429.9	114790
26	984.126021	333926464	0.008521	7276.9	115498
27	985.132433	227758208	0.008380	4962.7	117562
28	986.135614	73519288	0.008209	1600.7	120123
29	1007.120596	67884264	0.008835	1477.8	113996
30	1008.124636	45077780	0.008571	980.7	117615

Figure S4. Mass spectrum of HFHATN.

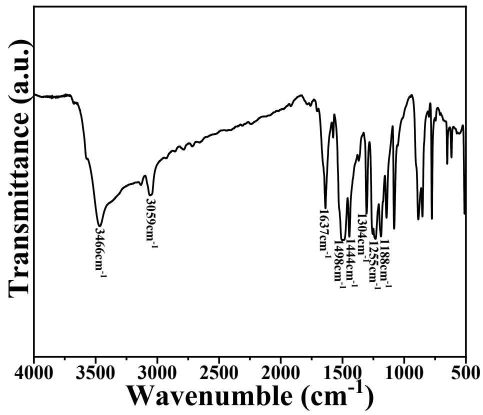


Figure S5. FTIR spectrum of HFHATN.

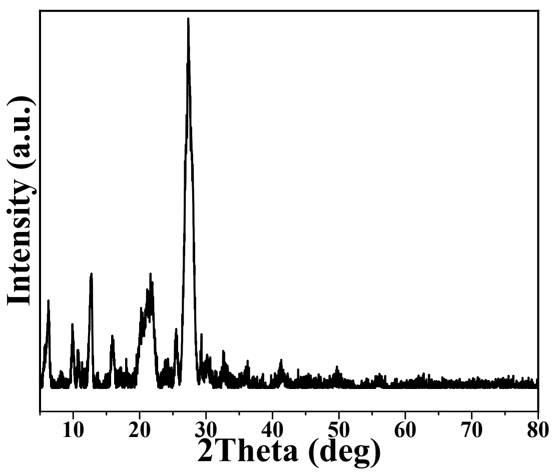


Figure S6. XRD spectrum of HFHATN.

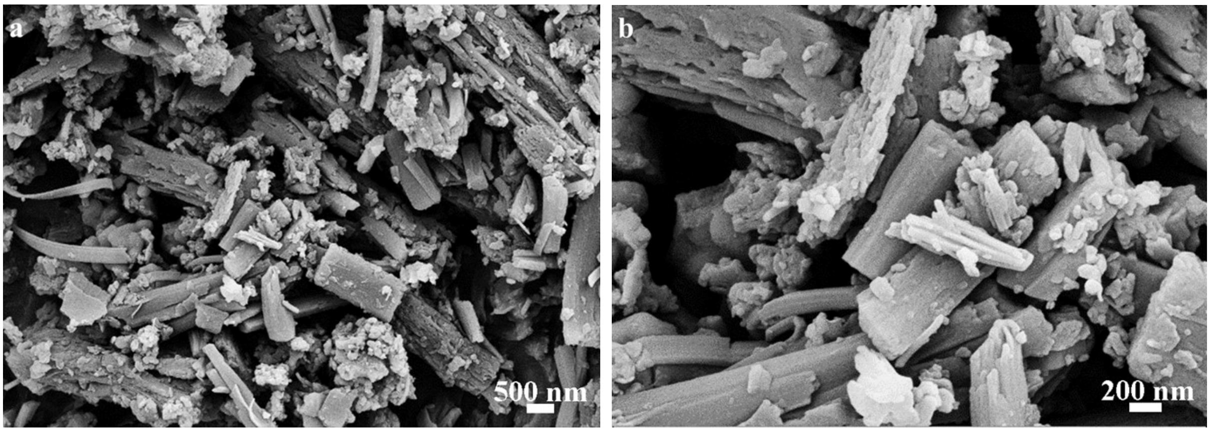


Figure S7. FESEM images of the HFHATN material (a) lower magnification (b) higher magnification.

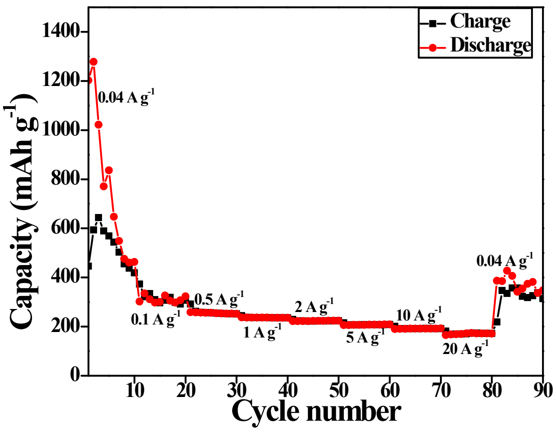


Figure S8. Rate capability of HFHATN electrode at various current densities.

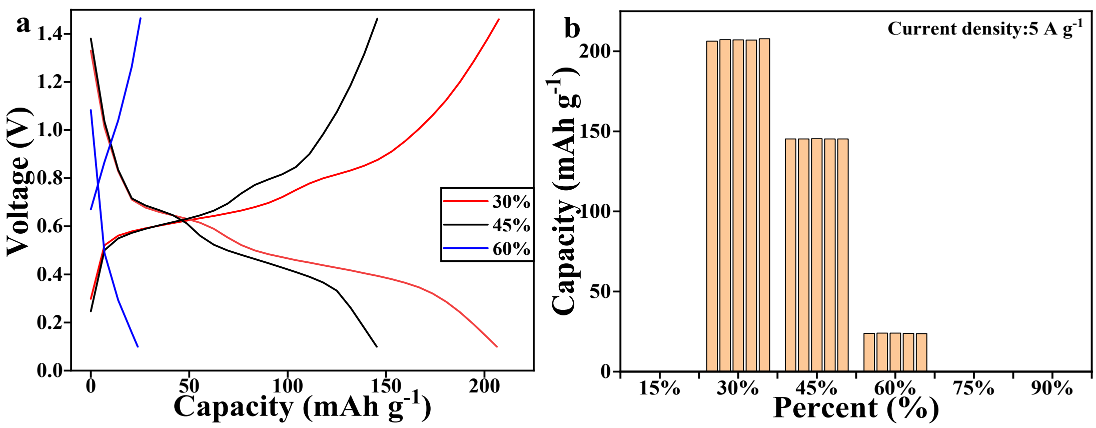


Figure S 9. Galvanostatic discharge/charge curves (a) and discharge histogram (b) of HFHATN electrode with different loading of active materials at a current density of 5A g^{-1}.

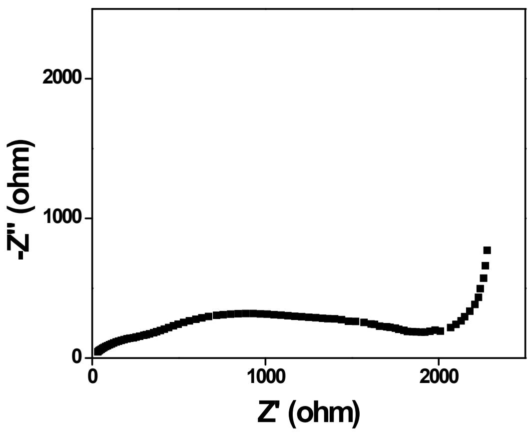


Figure S10. Nyquist plot of EIS measurement for the Zn//HFHATN battery.

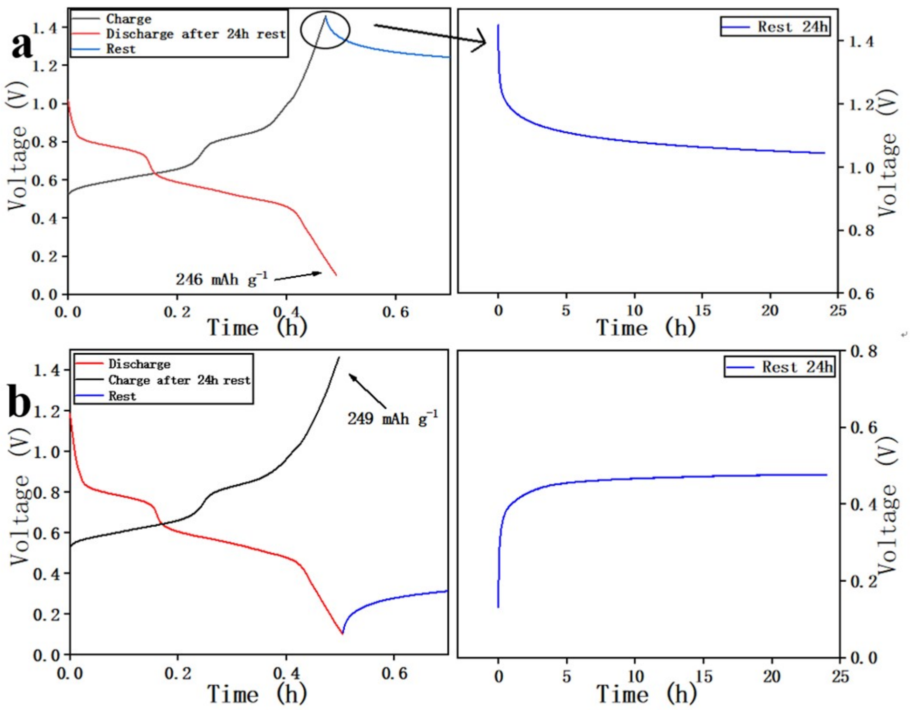


Figure S11. Self-discharge behavior tested of HFHATN electrode at a current density of 0.5A g^{-1}, (a) when firstly charged to 1.46 V, and then discharged to 0.1 V after rest for 24 hours, and (b) when firstly discharged to 0.1 V, and then charged to 1.46 V after rest for 24 hours

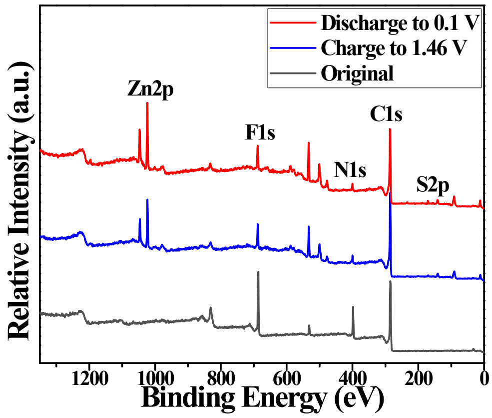


Figure S 12. Ex-situ XPS spectra of HFHATN electrode at pristine, fully discharged (0.1 V) and fully charged (1.46 V) states in a discharge/charge cycle. An aqueous coin battery was tested at a current density of 0.5 A g^{-1} for examination.

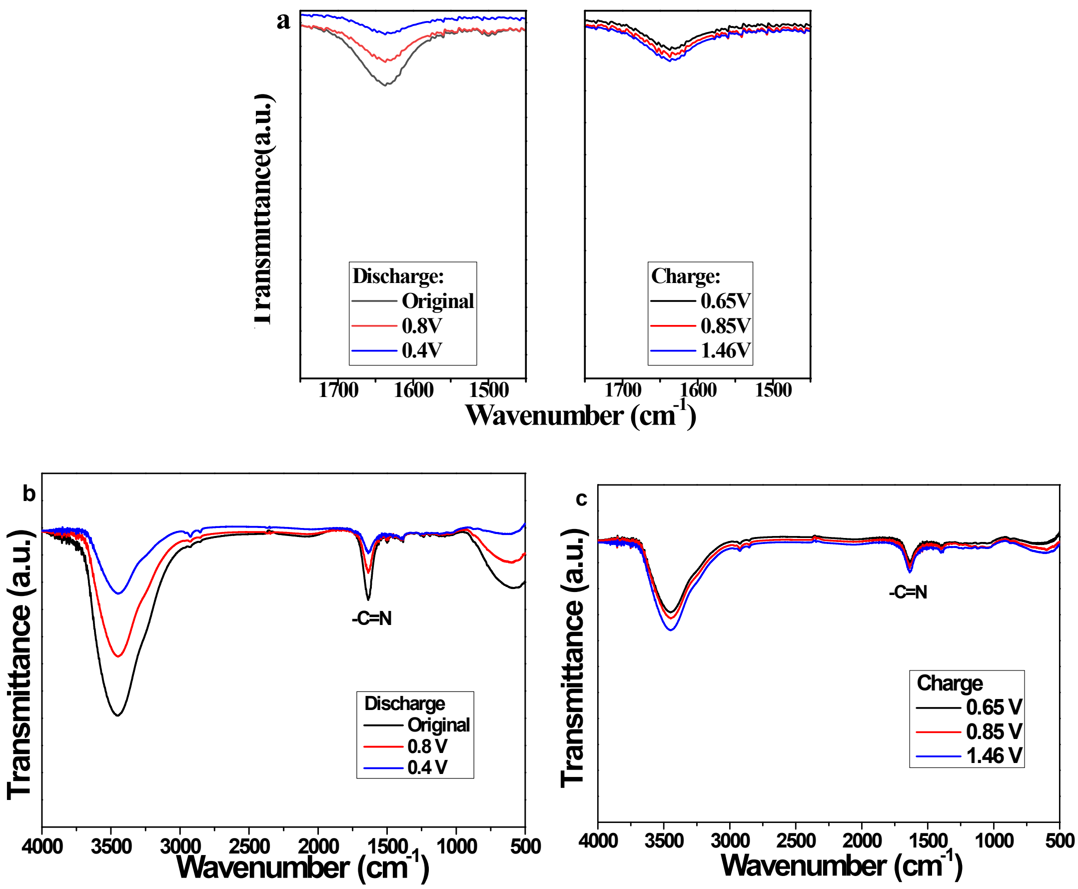


Figure S 13. Ex-situ FT-IR spectra of HFHATN electrode (a, b, c) at pristine, fully discharged (0.1 V) and fully charged (1.46 V) states in a discharge/charge cycle. An aqueous coin cell was tested at a current density of 0.5 A g^{-1} for examination.

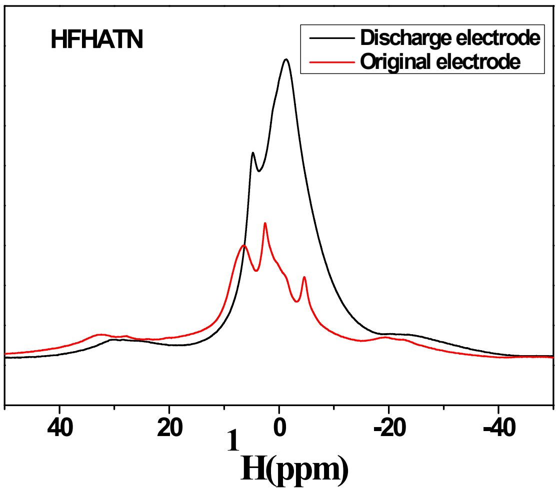


Figure S14. Solid-state ^{1}H NMR spectra of the HFHATN electrode under the original and discharge conditions

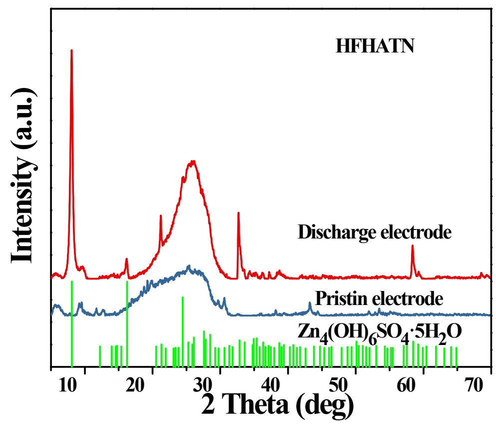


Figure S15. XRD patterns of the pristine electrode and the discharge electrode and $Zn_4(OH)_6SO_4 \cdot 5H_2O$ (PDF no 39-0688)

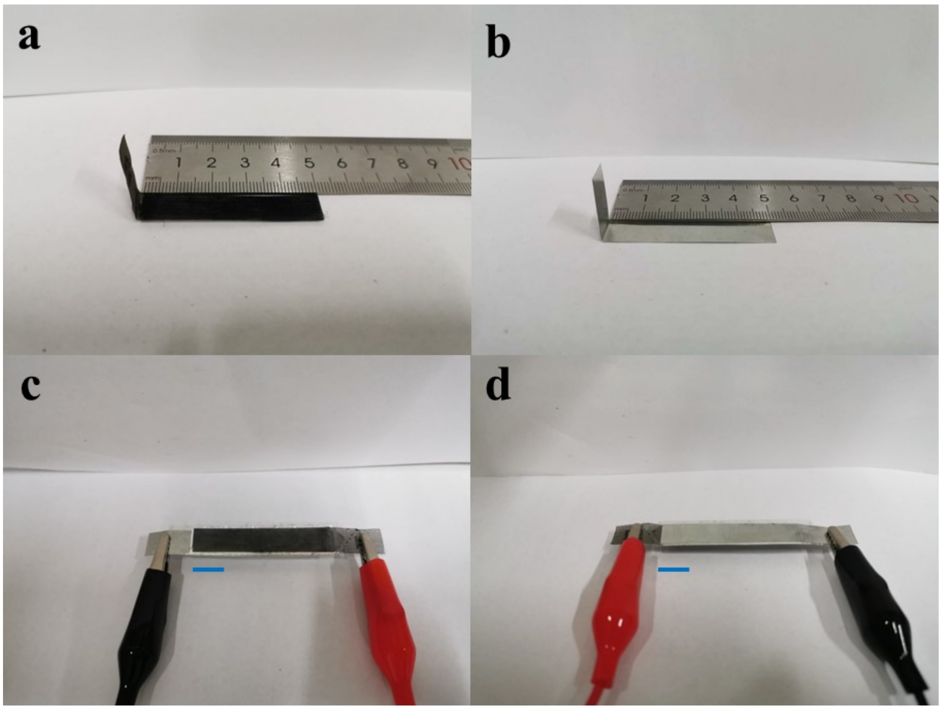


Figure S16. The images of the assembled flexible aqueous Zn//HFHATN battery. HFHATN cathode material (a) and Zn foil (b) in size of 1cm × 5 cm. The cathode-side-view (c) and the anode-side-view (d) of the flexible battery. (scale bar: 1cm)

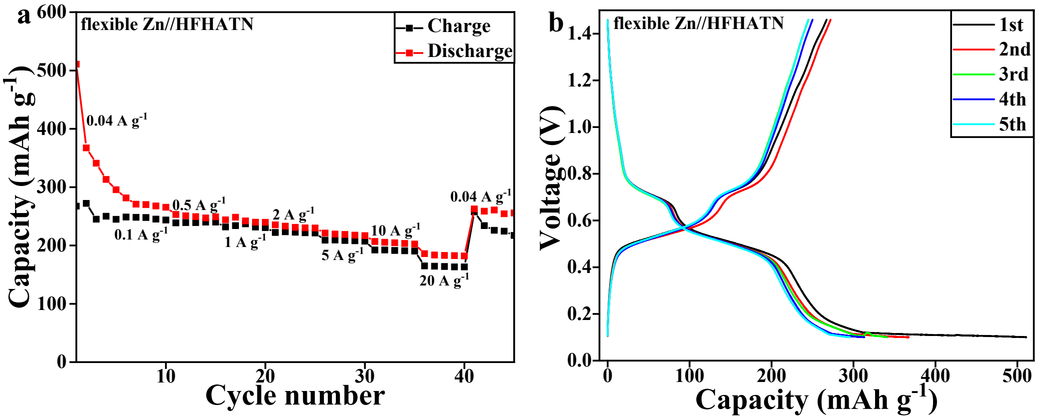


Figure S17. (a) The rate performances of flexible aqueous Zn//HFHATN battery. (b) The galvanostatic discharge/charge curves of flexible aqueous Zn//HFHATN battery at a current density of 0.04 A g^{-1}.

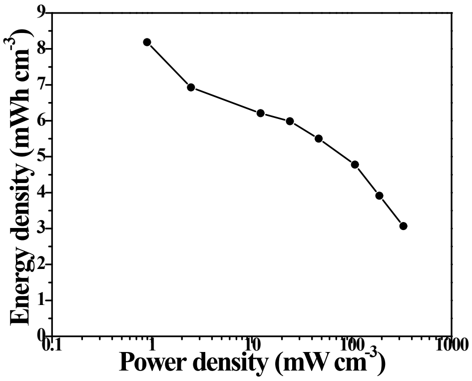


Figure S18. The Ragone plot of the flexible Zn//HFHATN battery.

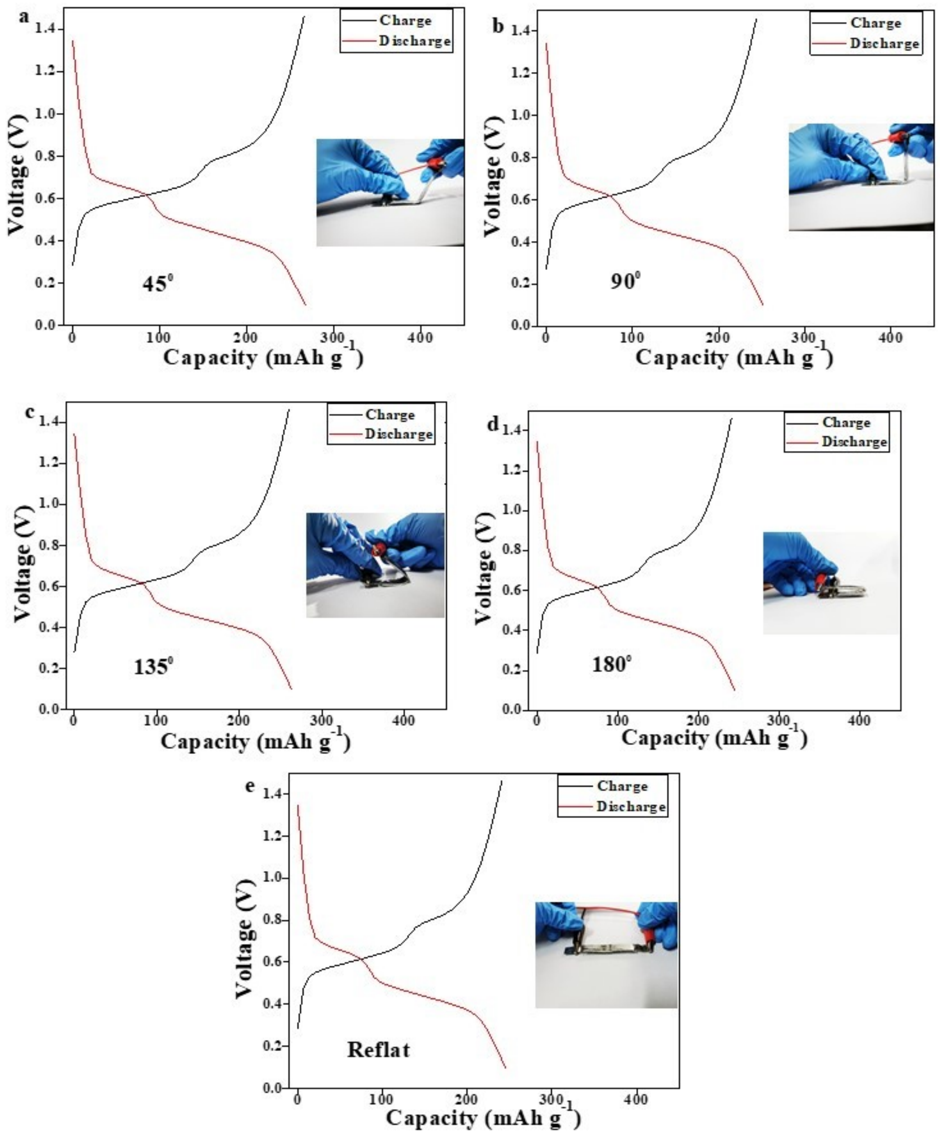


Figure S19 Flexible performance of the flexible Zn//HFHATN battery at bending degrees of (a) 45˚, (b) 90˚ (c) 135˚, (d) 180˚and (e) reflat. The insets are optical images of the flexible aqueous battery at corresponding bending degrees.

Table S1. Comparison of specific capacity in aqueous zinc batteries.

Electrode Materials	Specific capacity (Current density) /mA h g^{-1} (mA g^{-1})	Voltage / (V *vs* Zn/Zn^{2+})	Refs.
Quinone(C4Q)	333 (50)	0.8—1.3 V	24
Pyrene-4,5,9,10-tetraone	336 (40)	0.36—1.46V	25
p-chloranil	>200 (217)	0.8—1.4V	23
DTT	211/97 (50/2000)	0.3-1.4 V	27
NDI	~200/~25(0.2/10 C)	0.2-1.0 V	28
HATN	963/174 (40/6000)	0.1—1.46V	32
HMHATN	542/115 (40/8000)	0.1—1.46V	32
HATN	405/123 (100/20000)	0.3-1.1 V	33
HATN-3CN	313/190 (50/20000)	0.1-1.6 V	34
TCNAQ	166/55(50/1000)	0.6-1.8 V	35
PQ-Δ	225/210 (30/150)	0.25-1.6 V	29
PDB	205/176 (50 /1000)	0.2-1.55	30
PA-COF	265/68 (50/10000)	0.2-1.6 V	36
BDB	125 (25)	0.6-1.8 V	S1
PC/G-2	355/ 171 (50/5000)	0.2-1.9 V	S2
PANI	184/110 (20/10000)	0.5-1.6 V	S3
PANI	200/ 89 (50/5000)	0.5-1.5 V	S4
PDA	126.2/43.2(20/5000)	0.3-1.4 V	S5
PBQS	203/126 (20/1000)	0.2-1.8 V	S6
HqTp	276/125 (85/3750)	0.2-1.8 V	S7
HFHATN	461/172 (40/20000)	0.1-1.46 V	This work

Refs. 19- ---30 was given in the main-text

Table S2 Sum of electronic and zero-point energies in hartree calculated at the B3LYP/6-31+G (d, p) level.

	Energy/Hartree
Zn	-1779.1040434
H	-0.2151804
HFHATN	-1845.7542577
HFHATN_3Zn	-7183.21715873

1hartree =2625.5kJ/mol =27.21eV=627.51kcal/mol

Table S3. Comparison of energy and power densities between present work and previous reported flexible batteries and supercapacitors.

Flexible Materials	Energy Density ($mWh\ cm^{-3}$)	Power Density ($W\ cm^{-3}$)	Refs.
$HZnO@MnO_2$ SSC	0.04	0.00024	S8
MWCNT/CMF SSC	0.14	0.0027	S9
$H\text{-}TiO_2@MnO_2//H\text{-}TiO_2@C$ ASC	0.30	0.23	S10
Graphite/PANI SSC	0.32	0.054	S11
MVNN/CNT SSC	0.54	0.43	S12
$MnO_2//Fe_2O_3$ ASC	0.55	0.139	S13
VO_x//VN ASC	0.61	0.85	S14
Co_3O_4//graphene ASC	0.62	1.47	S15
ppy-coated paper SSC	1	0.27	S16
WO_{3-x}/MoO_{3-x}//PANI ASC	1.9	0.73	S17
$Ni(OH)_2$ NM//OMC fibers ASC	2.16	1.6	S18
PANI/Au ASC	10	3	S19
MWCNT//$Li4Ti_5O_{12}$ ASC	3.85	0.565	S20
ppy-MnO_2-CF SSC	6.16	0.4	S21
SWCNT/N-rGO microfibers SSC	6.3	1.085	S22
$LiMn_2O_4//Li_4Ti_5O_{12}$ planar battery	10	---	S23
$LiCoO_2$//LiPON//Li battery	2.2	---	S24
$LiMn_2O_4//Li_4Ti_5O_{12}$ wire battery	17.7	0.56	S25
$Na_{0.44}MnO_2//NaTi_2(PO_4)_3$ battery	23.8	3.8	S26
$Li_{1.1}Mn_2O_4//LiTi_2(PO_4)_3$ battery	124	11.1	S27
Zn//Ni-NiO@CF battery	0.67	---	S28
ZnO@CC//NiO@CC battery	7.76	0.54	S29

PTO//Zn battery	38.72	1.70	25
HATN//Zn battery	22.9	0.069	32
HMHATN//Zn battery	19.2	0.067	32
HFHATN//Zn battery	8.2	0.33	This work

Note: In the above table, *ASC* represents "asymmetric supercapacitor", and *SSC* represents "symmetric supercapacitor". In addition, Refs. 25 and 32 were given in the main-text. Refs. S1 to S29 are given in following Supplemental References section.

2. Note

2.1 Calculation about energy density and power density for Zn full batteries and flexible Zn batteries

In the main text, the current densities and specific capacities are calculated according to the mass of the active organic materials in the cathode. The detailed calculation about energy density and power density of Zn full batteries and flexible Zn batteries are presented as follows:

1. Calculation of the discharge capacity (mA h): The discharge specific capacity (mA h g^{-1}) multiplied by the mass of the active materials (m, g).

C = Specific capacity×m (1)

$$C = \text{Specific capacity} \times m \quad (1)$$

2. The calculation of the consumed zinc foil quality (g): The discharge capacity (mA h) was divided by the theoretical specific capacity of zinc (820 mA h/g).

$$m(Zn) = C/820 \quad (2)$$

3. Calculation of the energy density of Zn full/flexible battery is based following equation:

$$E = C \times V \times 1000/m \quad (3)$$

Where E, C, V and m stand for energy density (Wh kg^{-1}), the discharge capacity (mAh), the average discharge voltage (V) and the total mass of the cathode (active organic material) and anode (the consumed Zn) (kg), respectively.

4. Calculation of the power density of Zn full/flexible battery is based following equation:

P = E/t **(4)**

Where P is the power density (W kg^{-1}), E is the energy density (Wh kg^{-1}) and t is the discharge time (h).

5. Calculation of the volume energy density of flexible Zn battery is based following equation:

$$\mathbf{E_{vol} = C \times V/\nu} \quad \mathbf{(5)}$$

Where E_{vol}, C, V and ν stand for the volume energy density (mWhcm^{-3}), the discharge capacity (mAh), the average discharge voltage (V), and the volume of the flexible Zn battery (cm^3), respectively.

6. Calculation of the volume power density of flexible Zn battery is based following equation:

$$\mathbf{P = E_{vol}/t} \quad \mathbf{(6)}$$

Where P is the volume power density (mW cm^{-3}), E_{vol} is the volume energy density (mWh cm^{-3}) and t is the discharge time (h).

2.1 Calculation about the capacity contribution ratio from Zn^{2+} and H^+ based on inductively coupled plasma atomic emission spectroscopy (ICP-AES)

$Mass\ ratio\ of\ Zn\ and\ S = 53978.7 : 1681$

$$Mass\ ratio\ of\ Zn\ and\ S = \frac{53978.7}{65.39} : \frac{1682}{32.065}$$

$Molar\ ratio\ of\ coordinated\ Zn^{2+}\ and\ Zn_4(OH)_6SO_4{\cdot}H_2O$

$$= \left(\frac{53978.7}{65.39} - 4 * \frac{1682}{32.065}\right) : \frac{1682}{32.065}$$

$Molar\ ratio\ of\ coordinated\ Zn^{2+}\ and\ H^+$

$$= \left(\frac{53978.7}{65.39} - 4 * \frac{1682}{32.065}\right) : \left(6 * \frac{1682}{32.065}\right)$$

Capacity contribution $ratio$ from Zn^{2+} and H^+

$$= 2 * \left(\frac{53978.7}{65.39} - 4 * \frac{1682}{32.065}\right) : \left(6 * \frac{1682}{32.065}\right)$$

Based on above calculation, the capacity contribution from Zn^{2+} is about 79.7% and that from H^+ is about 20.3%.

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