Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting Information

Potassium Manganese Hexacyanoferrate With Improved Lifespan in $Zn(CF_3SO_3)_2$ Electrolyte for Aqueous Zinc-ion Batteries

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Table S1. Refinement results for KMHCF. Atomic and unit cell parameters from the least squares fitting routines in Fullprof (R_p = 7.47, R_{wp} = 9.63, χ^2 = 1.48).

P 2_1/n, a = 10.0912 Å, b = 7.3013 Å, c = 6.9716 Å, β = 90.2634°, V = 513.654					
Atom	**Wyck.**	**x/a**	**y/b**	**z/c**	**Occ.**
Fe1	2a	0	0	0	0.964
Mn1	2d	0	0.5	0.5	1
N1	4e	0.4514	0.2530	0.8542	0.964
C1	4e	0.5053	0.6386	0.2778	0.964
N2	4e	0.0525	0.3159	0.7426	0.964
C2	4e	0.9651	0.8058	0.1399	0.964
N3	4e	0.2077	0.4588	0.3700	0.964
C3	4e	0.7018	0.4837	0.5468	0.964
K1	4e	0.7387	0.9397	0.4726	0.911

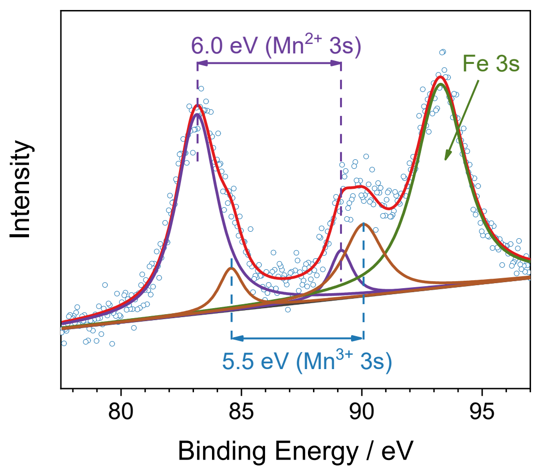

Figure S1. High resolution Mn 3s XPS results of KMHCF.

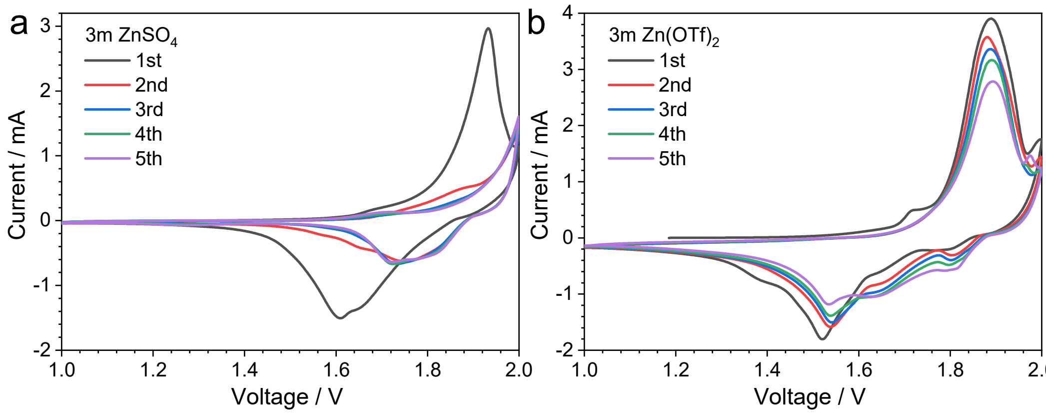

Figure S2. Cyclic voltammetry curves of KMHCF in zinc sulfate.

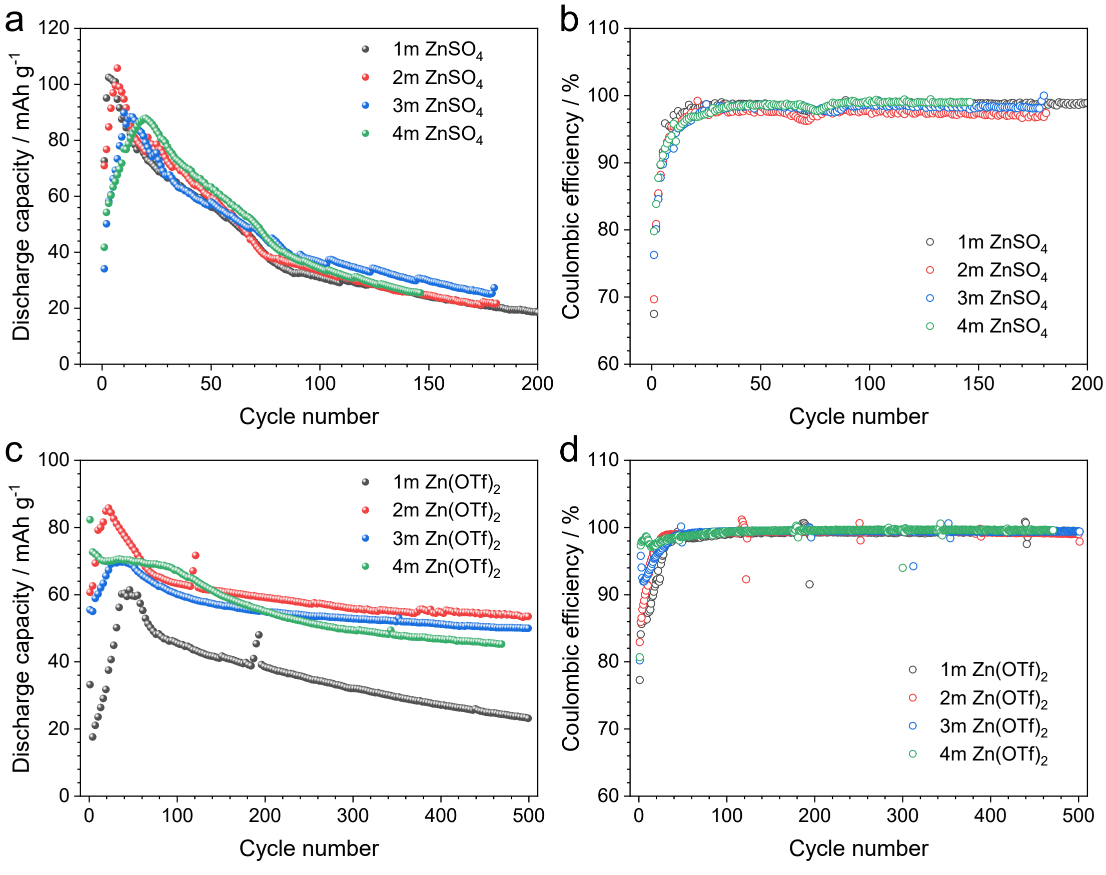

Figure S3. Long term cycle performance of KMHCF in $ZnSO_4$ (a, b) and $Zn(OTf)_2$ with concentration from 1m to 4m at a current density of 250 mA·g^{-1}

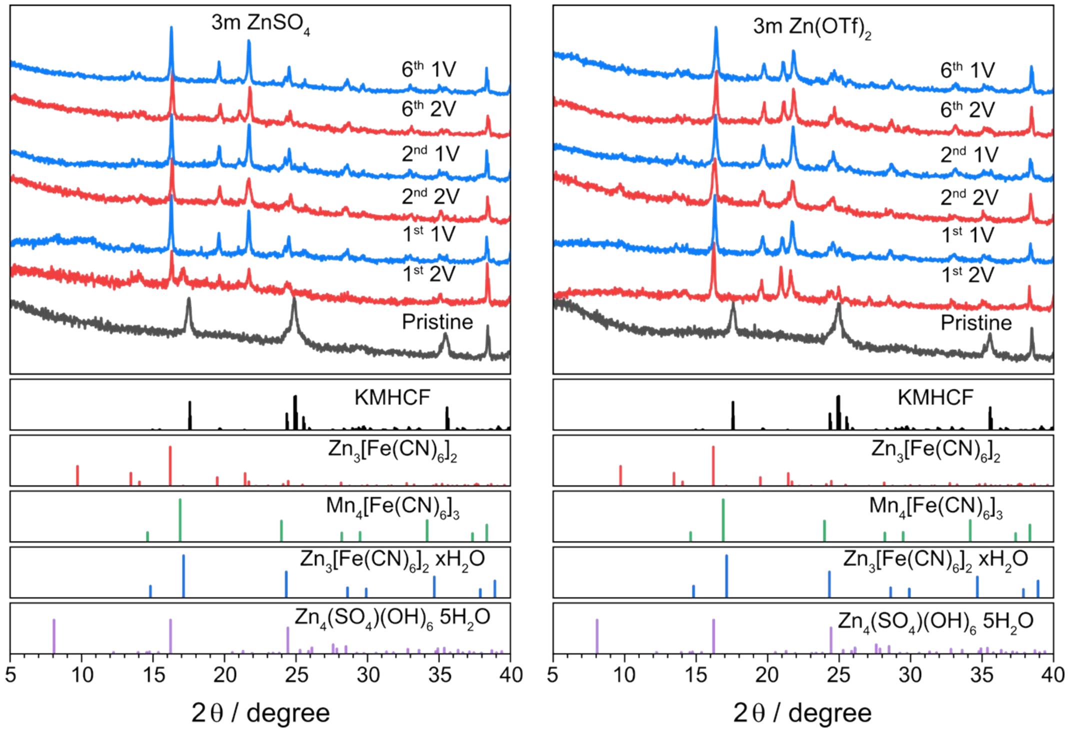

Figure S4. Ex-situ XRD patterns of the KMHCF electrodes at different states in $ZnSO_4$ and $Zn(OTf)_2$ electrolytes.

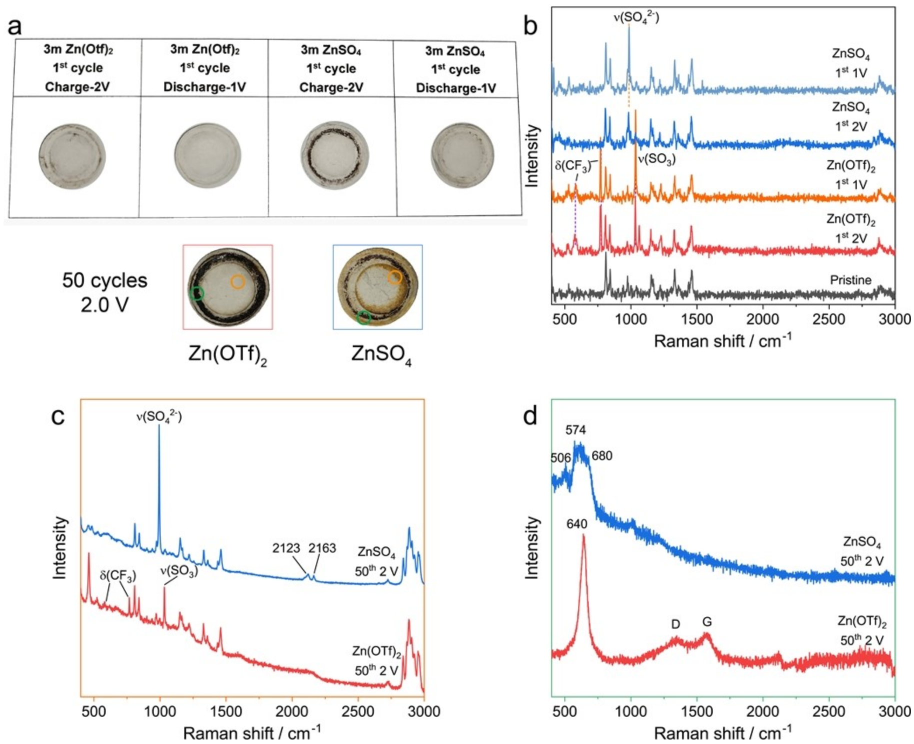

Figure S5. The photograph (a) and Raman spectra (b-d) of the separators after galvanostatic cycling in the KMHCF/Zn batteries with different electrolytes.

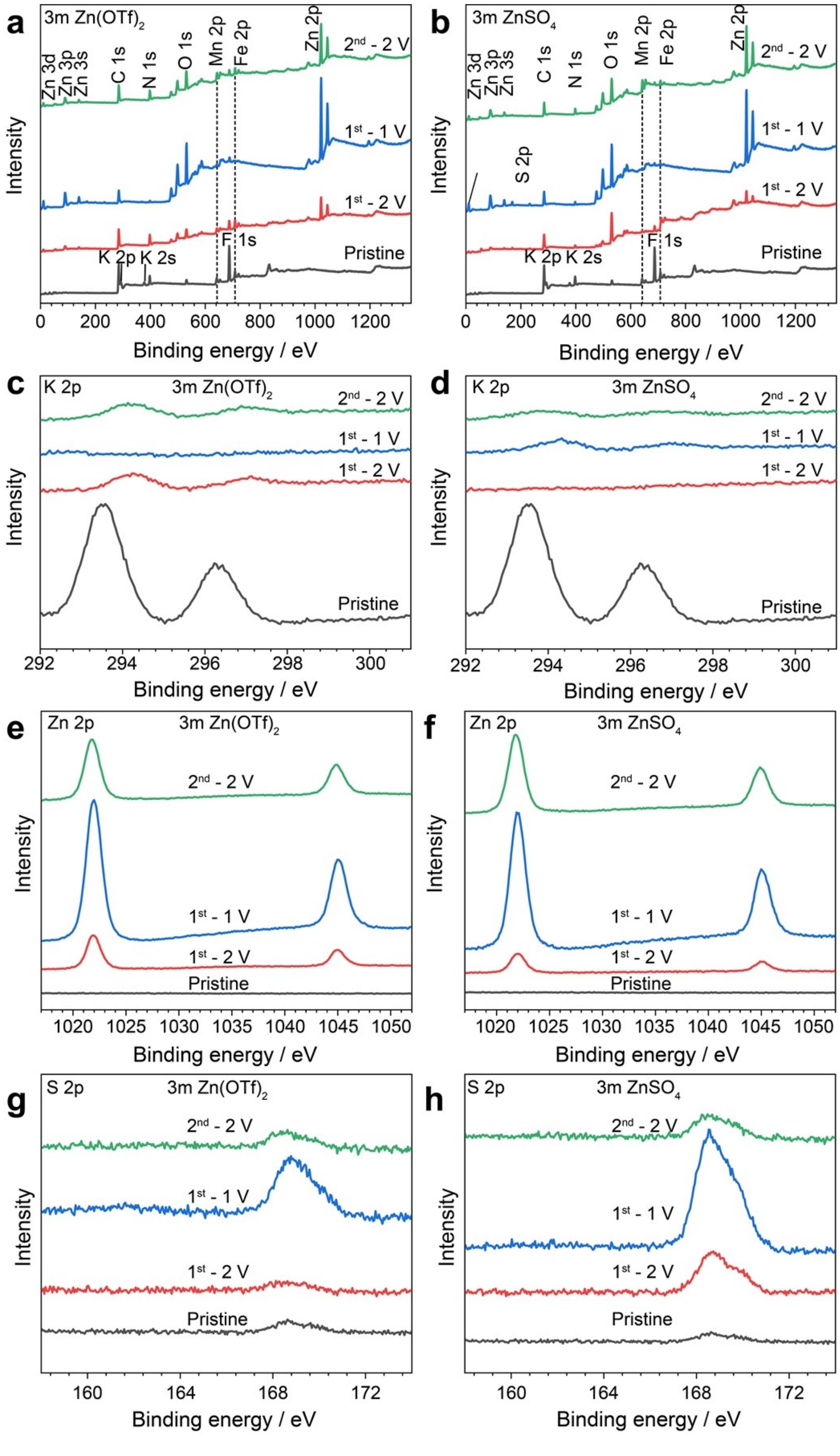

Figure S6. The XPS results of the KMHCF electrodes at different states in $ZnSO_4$ and $Zn(OTf)_2$ electrolyte

Table S2. Caparison of the electrochemical performance of various M-HCFs as cathode materials in ARZIBs

Cathode materials	**Electrolyte**	**Specific capacity at small current density**	**Long-term capacity retention**	**Reference**
ZnHCF	1M $ZnSO_4$	65.4 $mAh \cdot g^{-1}$ at 1C	76% retains after 100 cycles at 60 $mA \cdot g^{-1}$; 81% retains after 100 cycles at 300 $mA \cdot g^{-1}$	1
$KCuFe(CN)_6$	0.1M $Zn(ClO_4)_2$	~55 $mAh \cdot g^{-1}$	58.1% retains after 500 cycles	2
CuZn-HCF	0.02M $ZnSO_4$	<60 $mAh \cdot g^{-1}$ at 1C	85.54% after 1000 cycles at 1C	3
$CoFe(CN)_6$	4M $Zn(CF_3SO_3)_2$	173.4 $mAh \cdot g^{-1}$ at 300 $mA \cdot g^{-1}$	near 100% after 2200 cycles at 3.0 $A \cdot g^{-1}$	4
V-HCF	4M $Zn(CF_3SO_3)_2$	187 $mAh \cdot g^{-1}$ at 500 $mA \cdot g^{-1}$	96.5% retains after 500 cycles at 2.0 $A \cdot g^{-1}$	5
$K_{1.6}Mn_{1.2}Fe(CN)_6$	2 M $Zn(ClO_4)_2$		49% retains after 120 cycles at 200 $mA \cdot g^{-1}$ (aqueous)	6
KMHCF	3m $Zn(CF_3SO_3)_2$	123.8 $mAh \cdot g^{-1}$ at 50 $mA \cdot g^{-1}$	90% retains after 500 cycles	This work

Reference

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