Supporting Information

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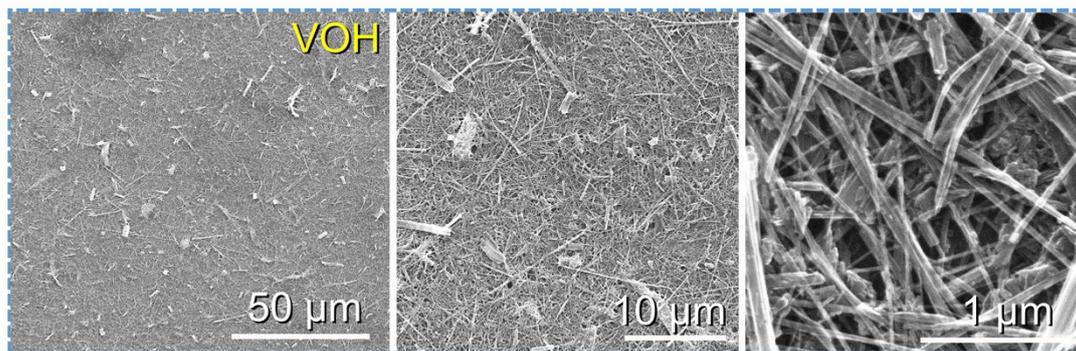
School of Physical Science and Technology, Lanzhou University, Lanzhou, Gansu 730000, China
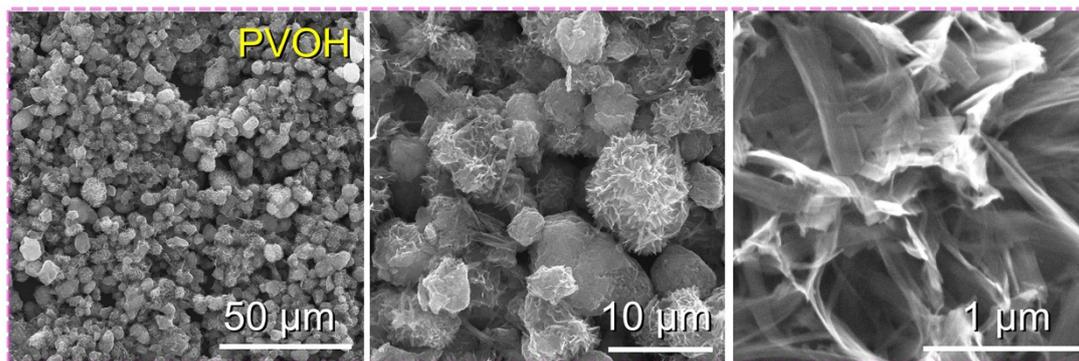
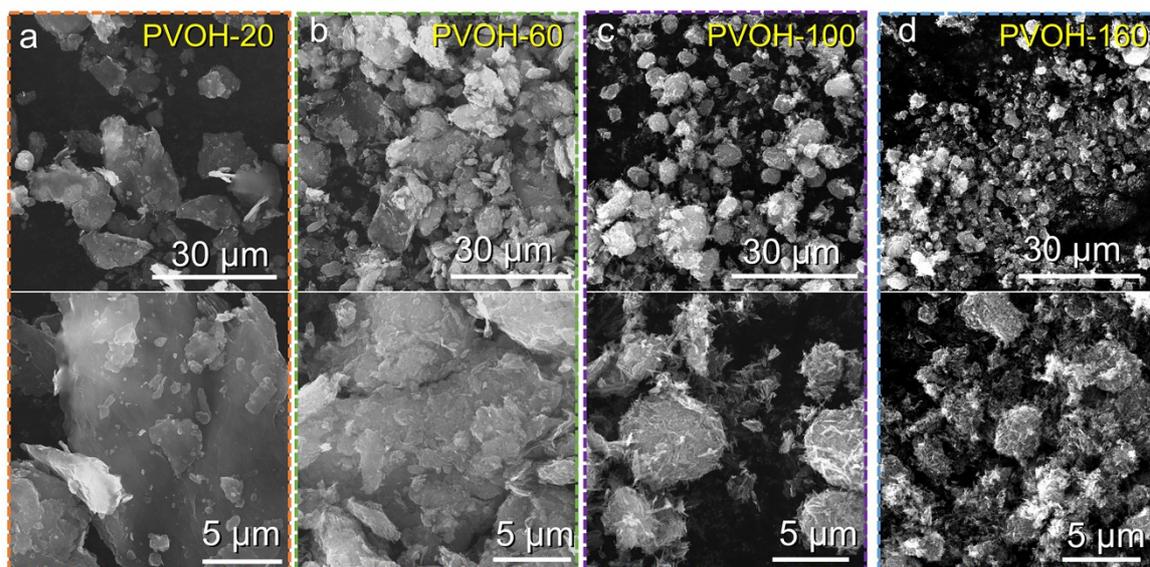* Corresponding authors: lanw@lzu.edu.cn (Wei Lan)

Fig. S1 Schematic illustration of the preparation process of VOH and PVOH.
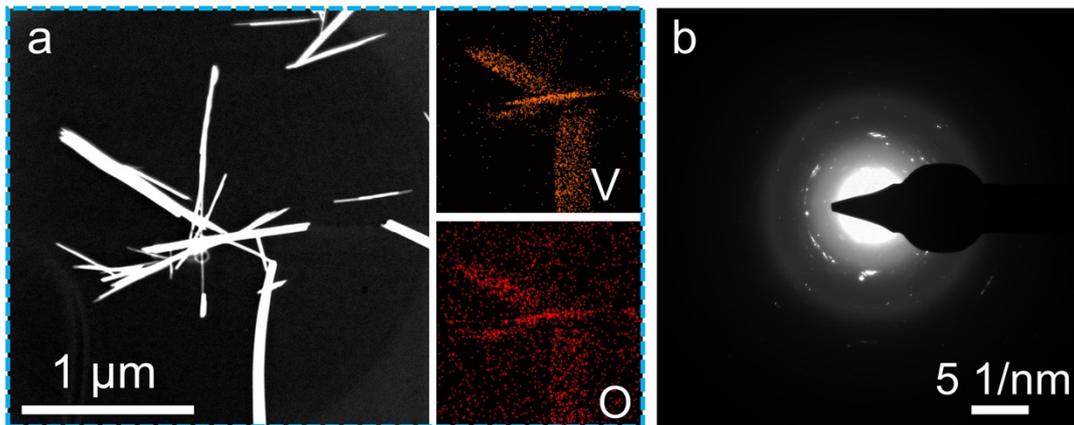
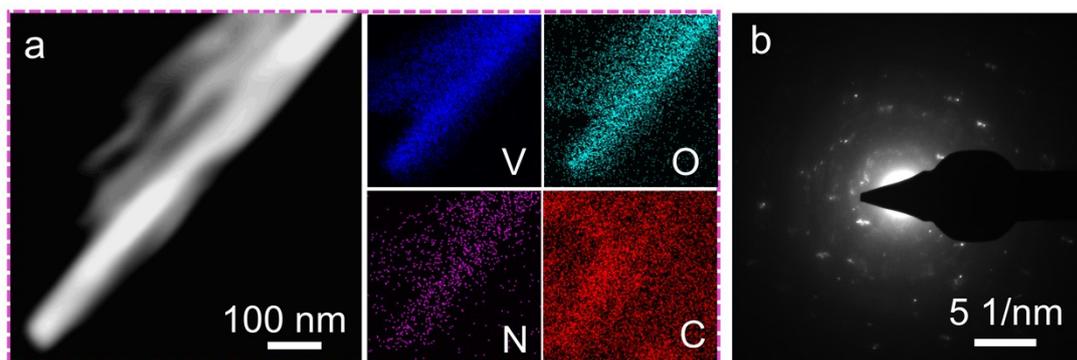**Fig. S2** SEM images of VOH at different magnifications.

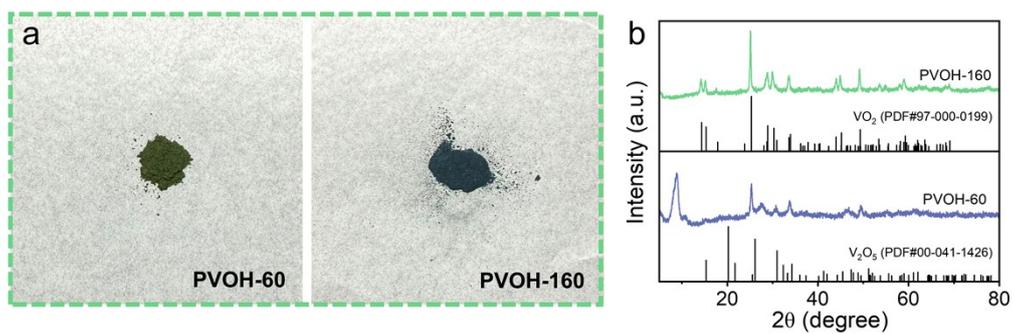**Fig. S3** SEM images of PVOH at different magnifications.

Fig. S4 SEM images at different magnifications of (a) PVOH-20, (b) PVOH-60, (c)PVOH-100, and (d) PVOH-160.
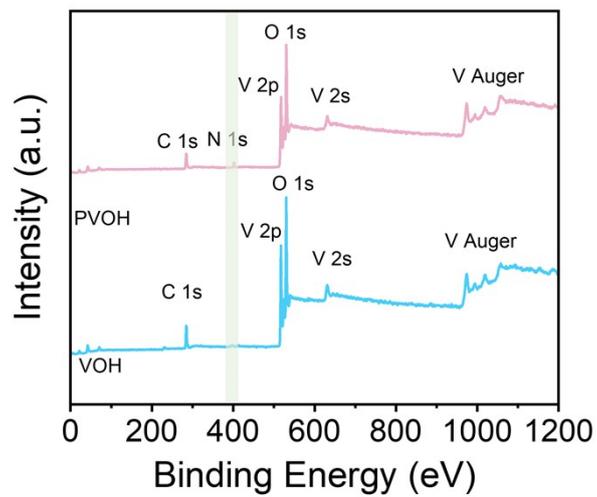
Fig. S5 (a) TEM element mapping images and (b) SAED image of VOH.
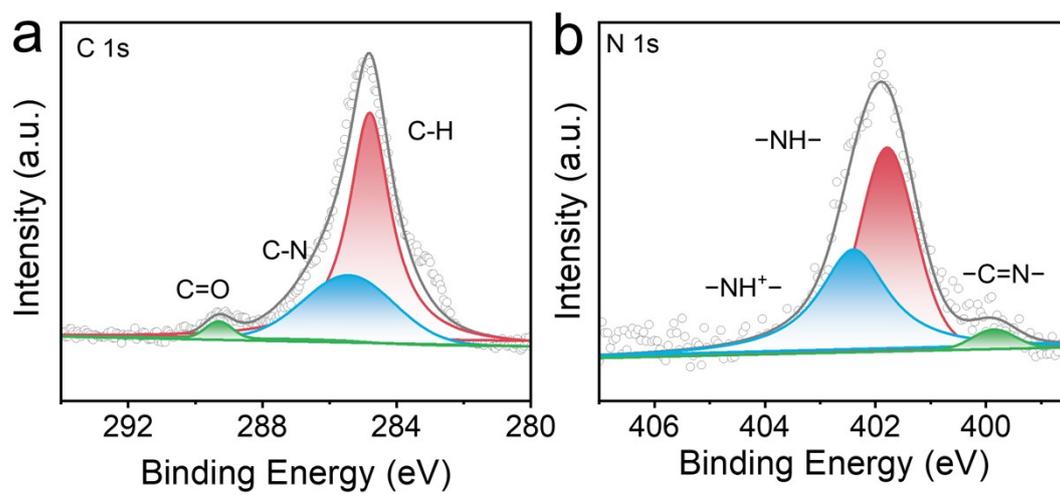
Fig. S6 (a) TEM element mapping images and (b) SAED image of PVOH.

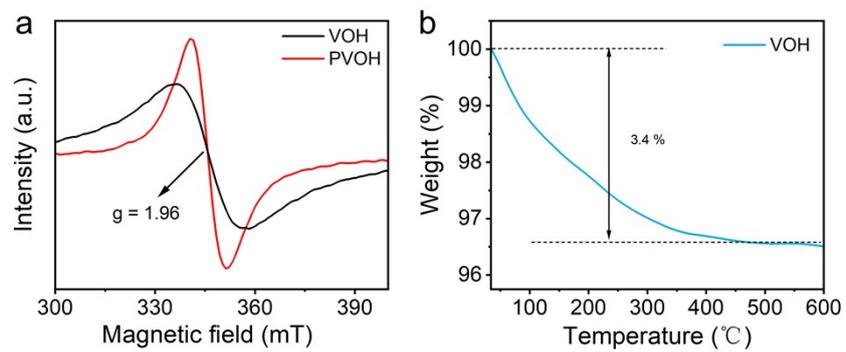**Fig. S7** (a) Optical photographs of PVOH-60 and PVOH-160 powder samples. (b)XRD patterns of PVOH-60 and PVOH-160.
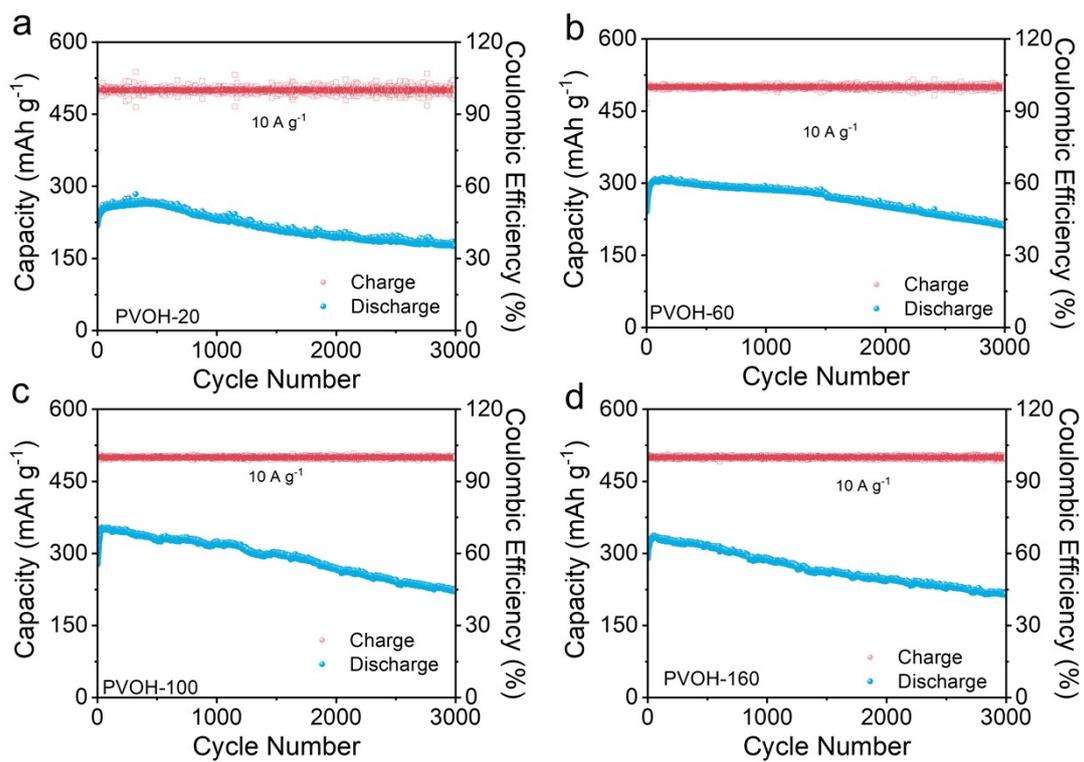
Fig. S8 XPS survey spectra of VOH and PVOH.

Fig. S9 The core-level XPS spectra of (a) C 1s and (b) N 1s of PVOH.

Fig. S10 (a) EPR spectra of VOH and PVOH . (b)TGA curves of VOH.

Fig. S11 Cycling performances of (a) PVOH-20, (b) PVOH-60, (c) PVOH-100, and (d) PVOH-160 cathodes at 10 A g⁻¹.

Table S1. Electrochemical performance of PVOH electrodes with different PPy

contents at 10 A g^{-1}

Cathode	Initial capacity (mAh g^{-1})	Capacity after 3000 cycles (mAh g^{-1})	Capacity retention (%)
PVOH-20	261.5	176.4	67.4
PVOH-60	307.8	212.1	68.9
PVOH-100	351.9	205.2	58.3
PVOH-160	331.1	216.7	65.4
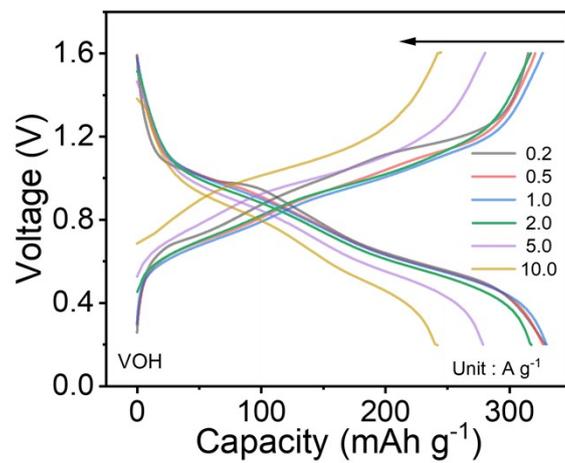
Fig. S12 Charge-discharge profiles of VOH at various current densities from 0.2 to 10 A g^{-1}.
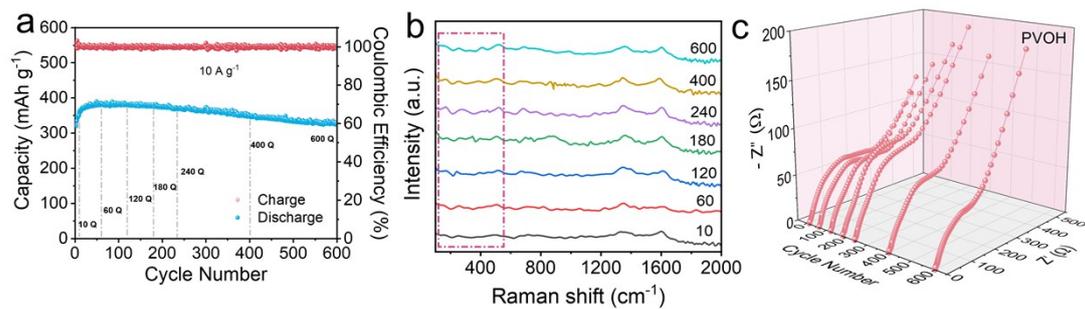
Fig. S13 (a) Cyclic performances of PVOH at 10 A g⁻¹ over 600 cycles. (b) Raman spectra of PVOH at selected cycle numbers. (c) EIS curves of PVOH during cycling.
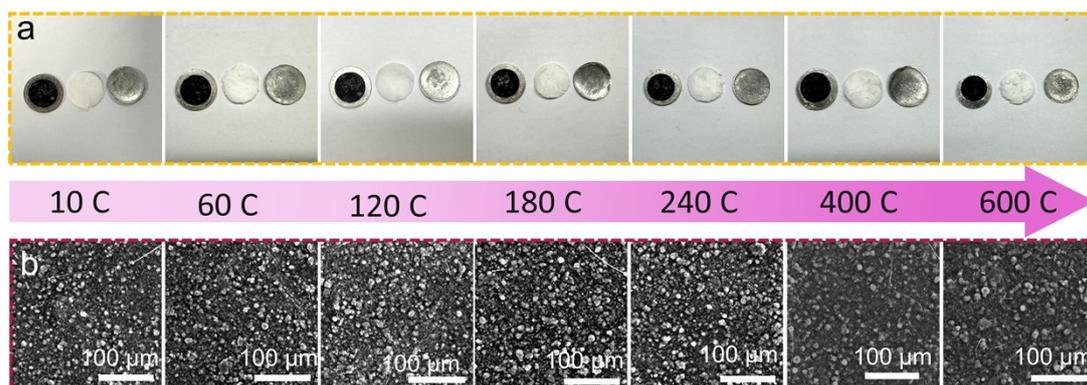
Fig. S14 Photographs of disassembled PVOH-based Zn-ion batteries (cathode, anode, and separator) after 10–600 cycles. (b) SEM images of the PVOH cathode surface after corresponding cycles.
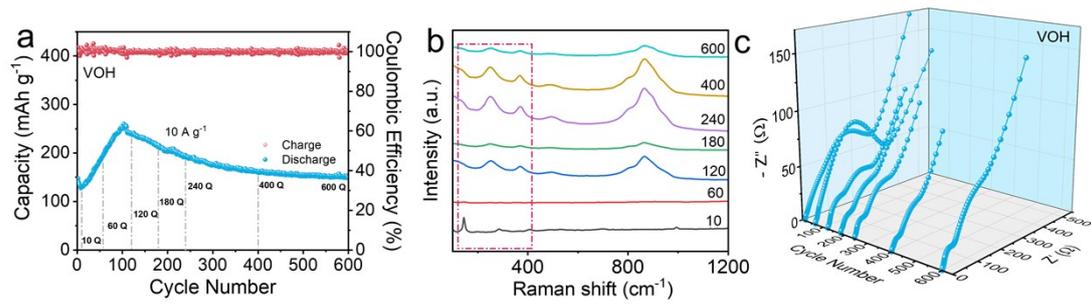
Fig. S15 (a) Cyclic performances of VOH at 10 A g⁻¹ over 600 cycles. (b) Raman spectra of VOH at selected cycle numbers. (c) EIS curves of VOH during cycling.

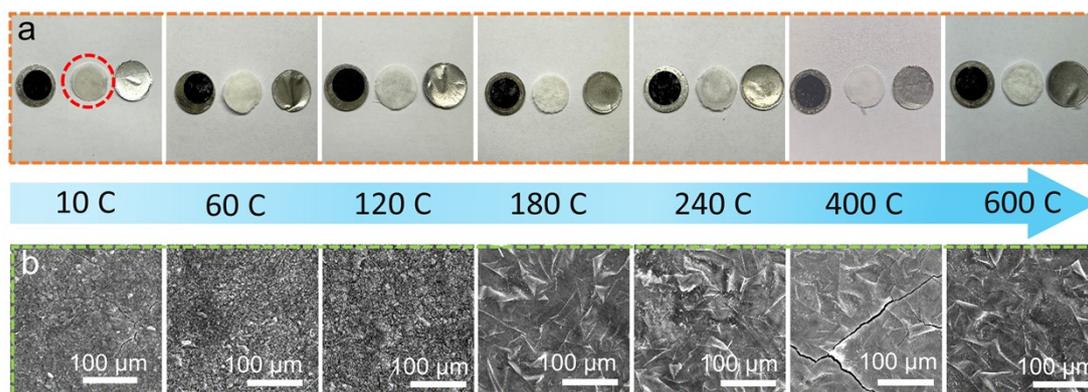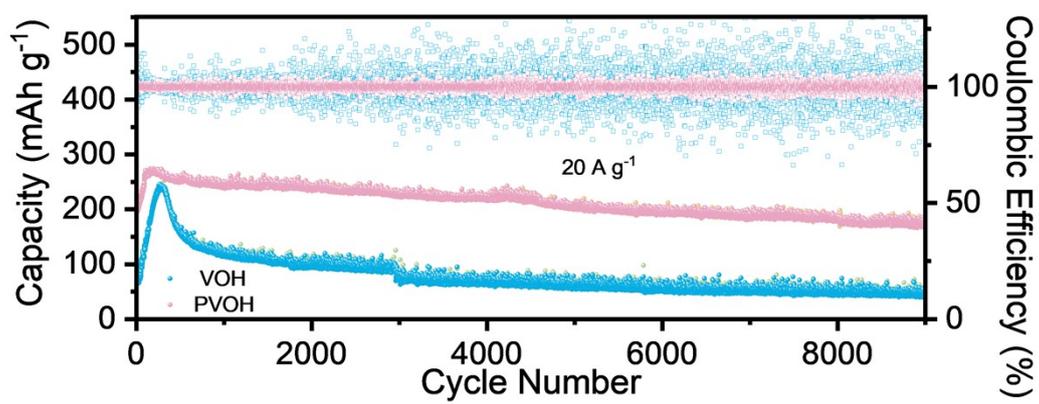**Fig. S16** (a) Photographs of disassembled VOH-based Zn-ion batteries (cathode, anode, and separator) after 10–600 cycles. (b) SEM images of the VOH cathode surface after corresponding cycles.
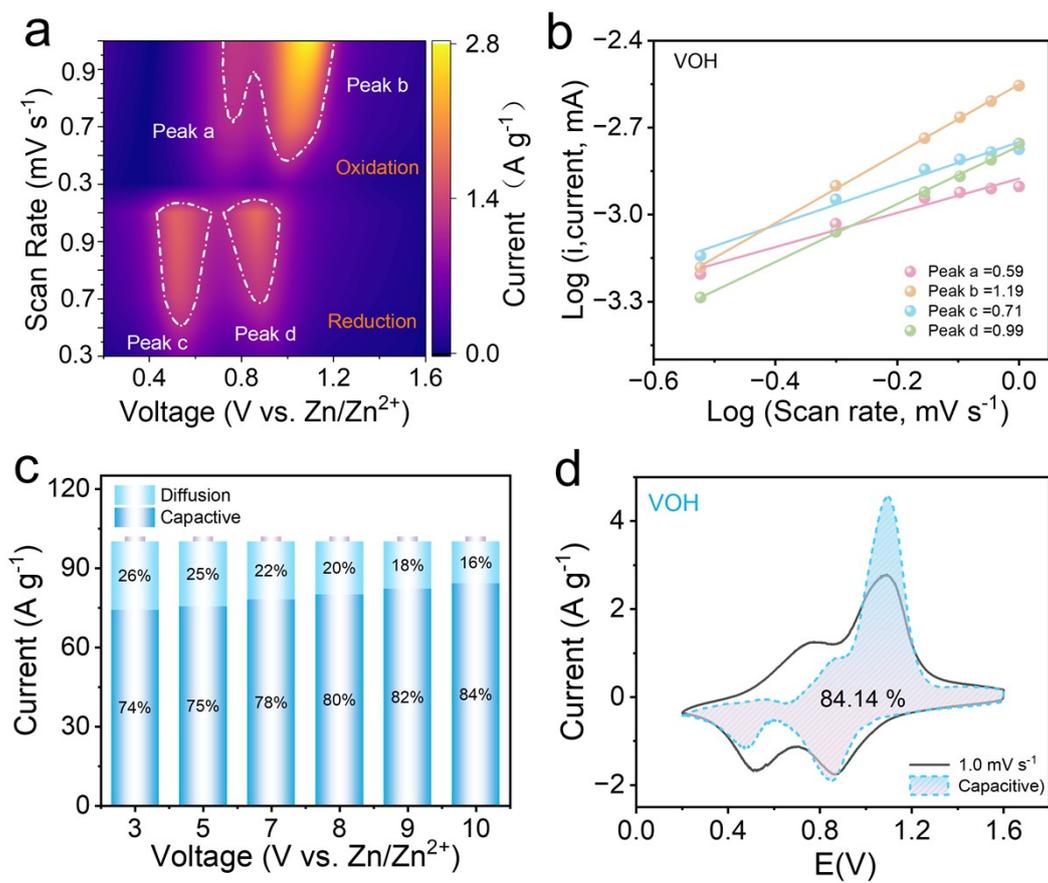
Fig. S17 Long-term cycling performances of VOH and PVOH at 20 A g^{-1}.

Fig. S18 (a) CV contour of VOH at various scan rates from 0.3 to 1.0 mV s^{-1}. (b) The b-value is based on the peak currents of the CV curves at various scan rates. (c) Capacitive contribution ratio at different scan rates. (d) CV curves with the capacitive contribution at 1 mV s^{-1}.

Table S2. Comparison of the overall performances with reported LDH-based electrodes.

Cathode	Voltage window (V)	Electrolyte	Capacity (mAh g^{-1}) /Current density (A g^{-1})	Capacity retention	Ref.
BTA-VOH	0.2-1.6	3 M Zn(CF$_3$SO$_3$)$_2$	408.0/0.5 291.0/8	92% (8 A g^{-1} after 3000 cycles)	[42]
O$_d$-HVO@PPy	0.2-1.6	2 M ZnSO$_4$	346.5/0.5 252.6/6	-	[29]
V$_6$O$_{13-x}$/rGO	0.2-1.5	3 M Zn(CF$_3$SO$_3$)$_2$	424.5/0.1 232.3/10	92% (5 A g^{-1} after 3000 cycles)	[43]
PVOH-M	0.4-1.7	3 M Zn(CF$_3$SO$_3$)$_2$	337.7/0.2 189.7/10	91% (10 A g^{-1} after 3000 cycles)	[26]
Zn$_{0.25}$V$_2$O$_5$	0.4-1.4	2 M ZnOTF + 1 mM NaOTF	293.0/0.05 167.0/2	83% (2 A g^{-1} after 2000 cycles)	[44]
CsVO	0.2-1.6	2 M Zn(CF$_3$SO$_3$)$_2$	400.0/0.2 224.0/20	99% (10 A g^{-1} after 5000 cycles)	[45]
NH$_4^+$-V$_2$O$_5$	0.2-1.8	3 M Zn(CF$_3$SO$_3$)$_2$	430.0/0.1 101.0/20	78% (10 A g^{-1} after 3600 cycles)	[46]
PANI100-V$_2$O$_5$	0.4-1.4	3 M Zn(CF$_3$SO$_3$)$_2$	360.0/0.5 216.0/10	75% (5 A g^{-1} after 2000 cycles)	[25]
VOH	0.2-1.6	3 M Zn(CF$_3$SO$_3$)$_2$	334/0.2 279/10	57% (10 A g^{-1} after 6000 cycles)	This work
PVOH	0.2-1.6	3 M Zn(CF$_3$SO$_3$)$_2$	439/0.2 351/10	70% (10 A g^{-1} after 7000 cycles)	This work