Supporting information

Interlayer Engineering by Ionic-Liquid Cation Pre-Intercalation for High-Performance Aqueous Zinc-Ion Batteries

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Notes

The capacitive and diffusion-controlled contributions were quantified using the following equation:

$$i(V) = k_1 v + k_2 v^{1/2}$$

where $i(V)$ represents the current response at a specific potential, and v denotes the scan rate. The term $k_1 v$ is associated with the capacitive-controlled process, whereas $k_2 v^{1/2}$ reflects the contribution governed by ion diffusion.[1]

The Zn^{2+} diffusion coefficient ($D_{Zn^{2+}}$) was estimated from the GITT measurements according to:

$$D_{Zn^{2+}} = \frac{4}{\pi\tau}\left(\frac{m_B V_M}{M_B A}\right)^2\left(\frac{\Delta E_s}{\Delta E_\tau}\right)^2$$

where τ denotes the duration of the current pulse, V_M and M_B are the molar volume and molar mass of the active material, respectively, and m_B represents the mass of active material. A is the effective electrode/electrolyte contact area. ΔE_s refers to the steady-state voltage variation between two consecutive relaxation periods, whereas ΔE_τ represents the voltage response during the corresponding current pulse.[2]

The $D_{Zn^{2+}}$ can be evaluated by the following equation:

$$D_{Zn^{2+}} = \frac{R^2T^2}{2A^2n^4F^4C^2\sigma_\omega{}^2}$$

where R is the gas constant, T is the absolute temperature, A is the area, n is number of electrons transferred per molecule, F is the Faraday constant, C is Zn^{2+} concentration, and σ_ω is the Warburg coefficient.[3]

Theoretical calculation

All first-principles calculations were performed using the Vienna Ab initio Simulation Package (VASP).[4, 5] The exchange-correlation interactions were treated within the generalized gradient

approximation (GGA) using the Perdew-Burke-Ernzerhof (PBE) functional, while the electron-ion interactions were described by the projector augmented-wave (PAW) method.[6, 7] A plane-wave energy cutoff of 400 eV was adopted, and Brillouin-zone integrations were sampled using a 1×2×1 Monkhorst-Pack k-point mesh. During structural relaxation, the electronic energy and atomic forces were converged to 1.0×10^{-5} eV and 0.05 eV $Å^{-1}$, respectively. Long-range van der Waals interactions were incorporated using the DFT-D3 correction method.[8]

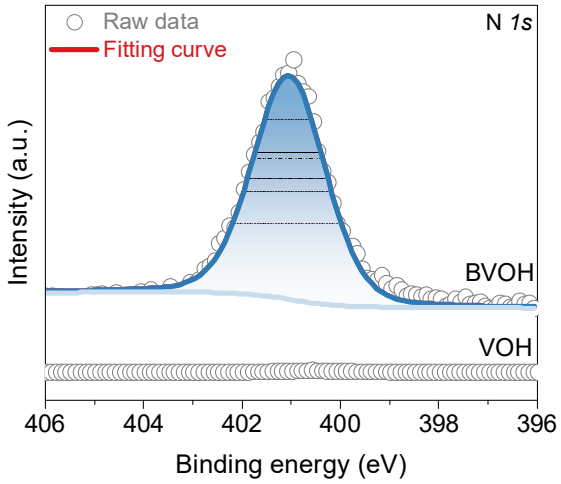


Figure S1. N 1s XPS spectra of VOH and BVOH.

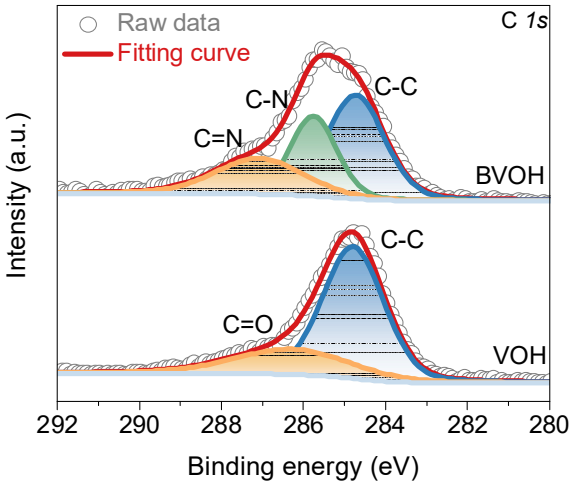


Figure S2. C 1s XPS spectra of VOH and BVOH.

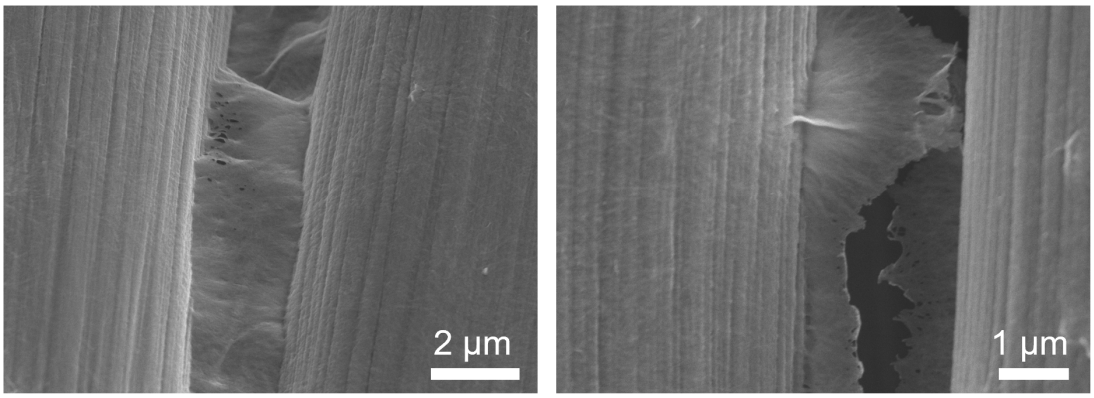


Figure S3. SEM images of VOH.

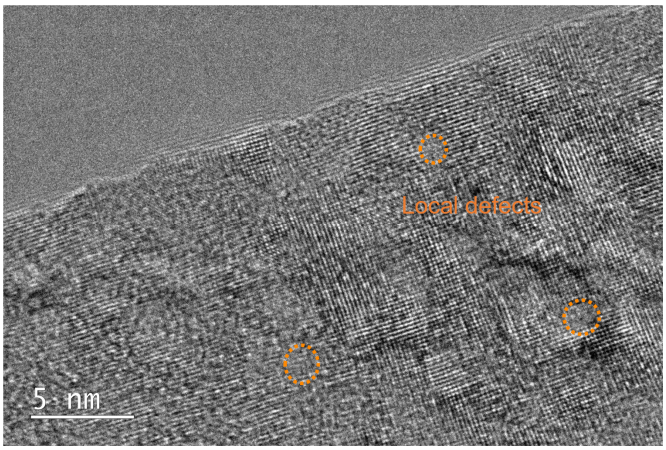


Figure S4. HRTEM image of BVOH.

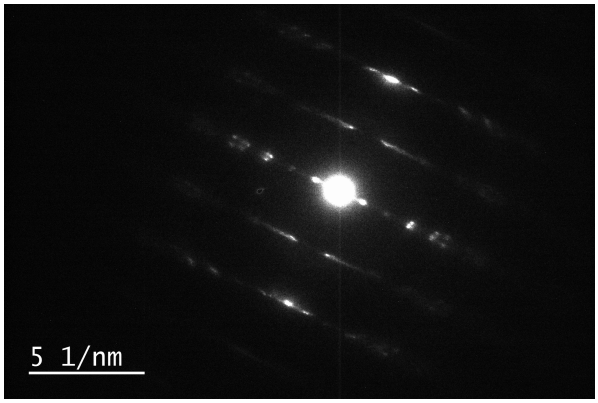


Figure S5. SAED image of BVOH.

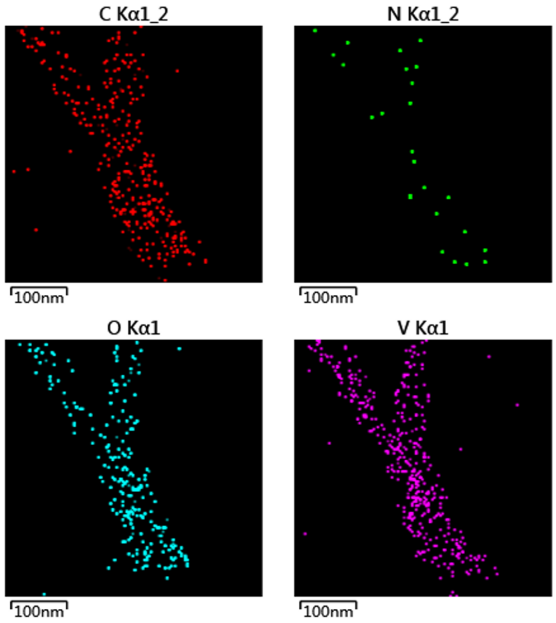


Figure S6. EDS mappings of BVOH.

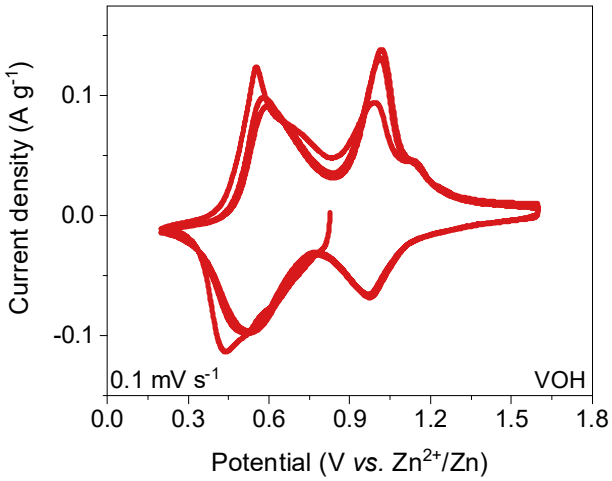


Figure S7. CV curves of VOH at 0.1 mV s^{-1}.

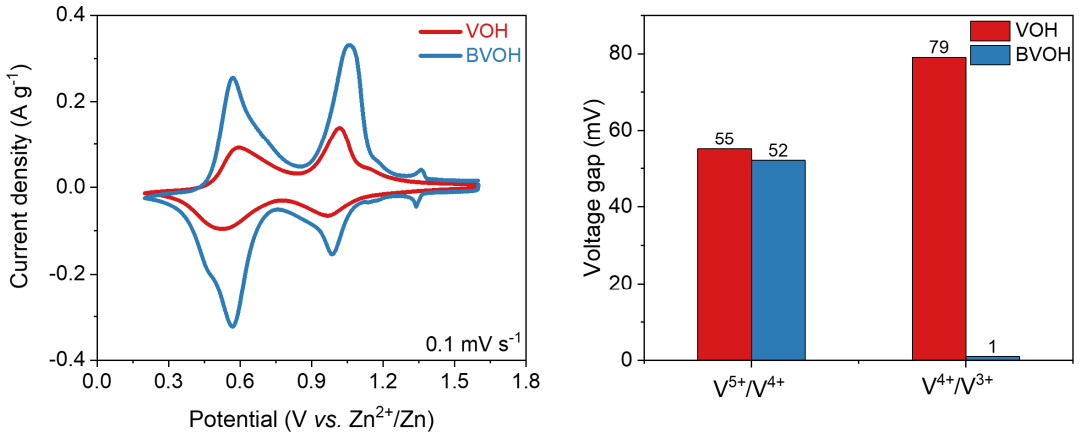


Figure S8. Compared CV curves and voltage gaps.

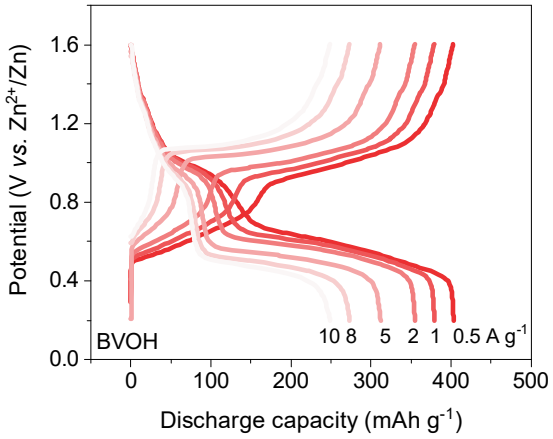


Figure S9. GCD curves of BVOH at various current densities.



Figure S10. Contact angle of VOH and BVOH.

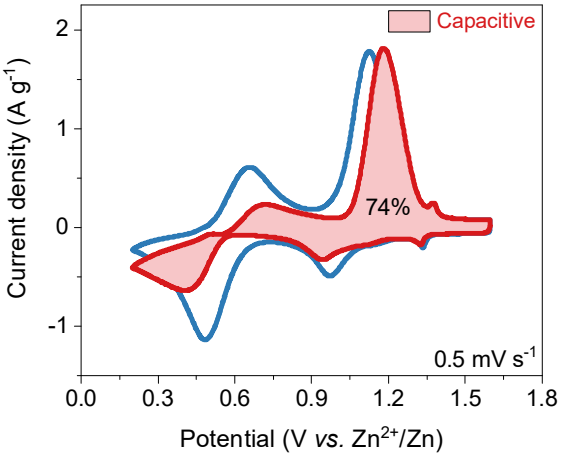


Figure S11. Capacitive contribution of BVOH at 0.5 mV s^{-1}.

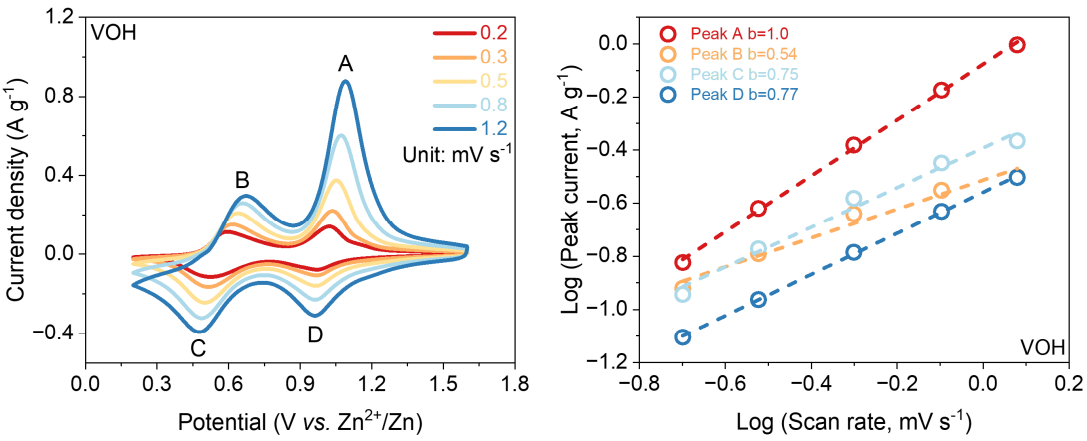


Figure S12. b values of VOH.

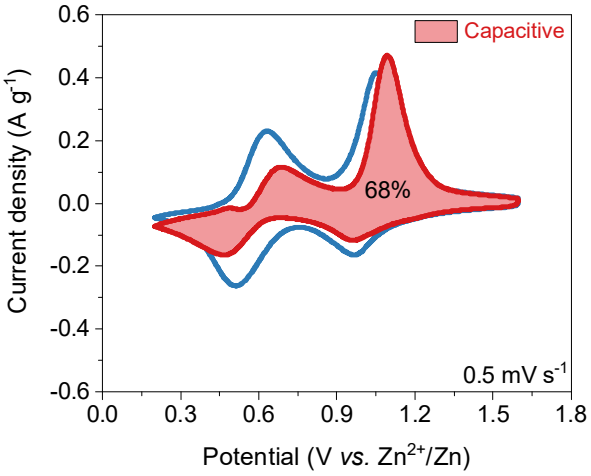


Figure S13. Capacitive contribution of VOH at 0.5 mV s^{-1}.

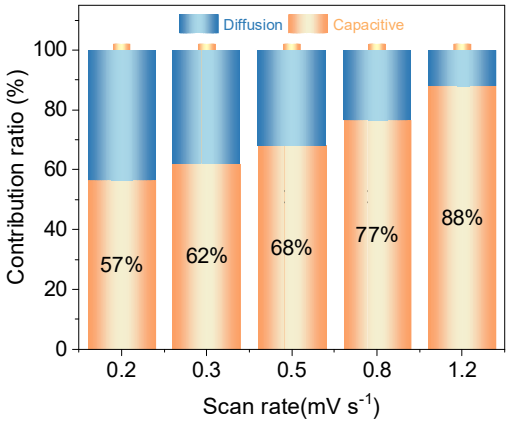


Figure S14. Capacitive contribution of VOH at various scan rates.

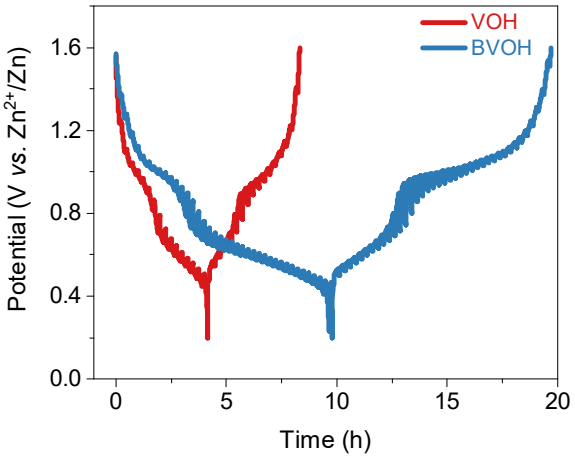


Figure S15. GITT curves of VOH and BVOH.

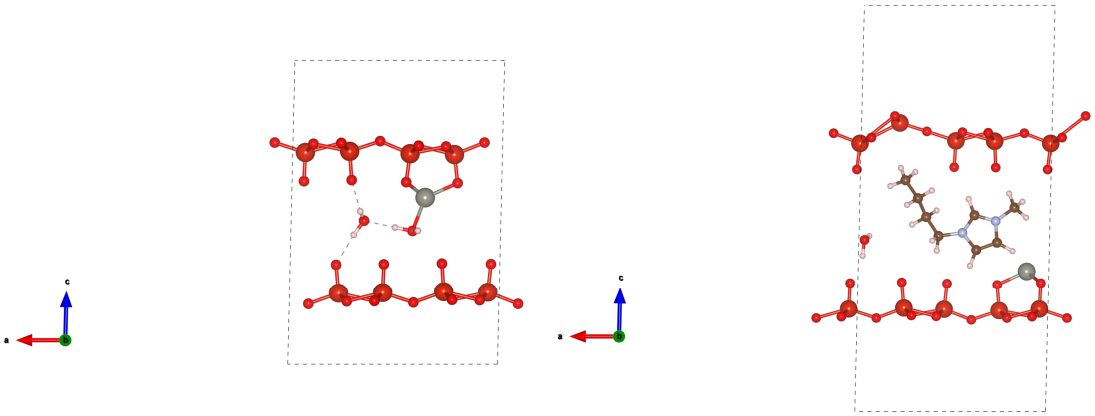


Figure S16. Calculated models of VOH and BVOH.

Table S1. Electrochemical properties comparison of BVOH with other organic species pre-intercalated vanadium-based cathodes.

Materials	Capacity	Rate capability	Cycling performance	Ref.
BVOH	452 mAh g^{-1} at 0.1 A g^{-1}	247 mAh g^{-1} at 10 A g^{-1}	90.3% capacity retention after 8000 cycles at 8 A g^{-1}	This work
Phen-VOH	386.5 mAh g^{-1} at 0.5 A g^{-1}	218.9 mAh g^{-1} at 10 A g^{-1}	81.4% capacity retention after 10000 cycles at 10 A g^{-1}	[9]
TNVO@GO	438.2 mAh g^{-1} at 0.2 A g^{-1}	318.5 mAh g^{-1} at 6 A g^{-1}	84.6% capacity retention after 3000 cycles at 6 A g^{-1}	[10]
EG-NVO	517 mAh g^{-1} at 0.5 A g^{-1}	203 mAh g^{-1} at 10 A g^{-1}	91% capacity retention after 1000 cycles at 10 A g^{-1}	[11]
PEO-LVO	438 mAh g^{-1} at 0.1 A g^{-1}	181 mAh g^{-1} at 10 A g^{-1}	90.5% capacity retention after 3000 cycles at 10 A g^{-1}	[12]
NVO/PoPDA@GO	433 mAh g^{-1} at 0.5 A g^{-1}	180 mAh g^{-1} at 5 A g^{-1}	74.9% capacity retention after 1000 cycles at 5 A g^{-1}	[13]
HNVO-Glu	401 mAh g^{-1} at 0.2 A g^{-1}	230 mAh g^{-1} at 10 A g^{-1}	87.2% capacity retention after 12000 cycles at 10 A g^{-1}	[14]
VO@pAP	383 mAh g^{-1} at 0.2 A g^{-1}	139 mAh g^{-1} at 5 A g^{-1}	82% capacity retention after 2100 cycles at 3 A g^{-1}	[15]
TMPA-VOH	403 mAh g^{-1} at 0.5 A g^{-1}	294 mAh g^{-1} at 8 A g^{-1}	87% capacity retention after 2000 cycles at 4 A g^{-1}	[16]

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