

Electronic supplementary information

An amphoteric betaine electrolyte additive enabling a stable Zn metal anode for aqueous batteries

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Experimental Section

Material synthesis

VO₂ was obtained by microwave assisted hydrothermal reaction (Monowave 200 synthesis system Anton Paar). 0.4 g of V₂O₅ and 0.6 g of H₂C₂O₄ · 2H₂O were dissolved in deionized water. In a microwave reactor, the solution was heated at 75 °C for 5 min, and then at 180 °C for 10 min. After cooling to room temperature, the precipitation was filtered, washed with deionized water and ethanol, and dried at 60 °C in a vacuum oven.

Characterizations

¹H nuclear magnetic resonance (¹H NMR) was carried out on a Bruker Avance NEO 600 M. Fourier transform infrared (FT-IR) spectroscopy was collected on the Lambda 650S spectrometer (UK PerkinElmer). Raman spectra were obtained from a LabRAM HR Evolution series high-resolution Raman spectrometer (Nano Wizard Ultra Speed & inVia Raman, Germany) with a laser wavelength of 633 nm. Zeta potentials were measured on Malvern Zetasizer Nano S90. X-ray diffraction (XRD) was performed on a PANalytical Empyrean diffractometer with Cu-Kα radiation. Morphologies were examined using field-emission scanning electron microscope (HITACHI, SU8010, Japan).

Electrochemical measurements

VO₂ electrodes were prepared by mixing VO₂ with Ketjen Black (KB) and polyvinylidene fluoride (PVDF) at 7:2:1 weight ratio in N-methyl-2-pyrrolidone (NMP). Zn//Zn cells and Zn//Cu cells were assembled in CR2032 coin cells. Zn//VO₂ cells were assembled in PFA Swagelok cells. Cyclic voltammetry (CV) and linear polarization tests were carried out in T-shaped three-electrode PFA Swagelok cells. Linear polarization was carried out with Zn, graphite paper and SCE as the working, counter and reference electrodes, respectively. The electric double-layer capacitance (EDLC) was measured with Zn as the working, counter and reference electrodes. The EDLC was calculated according to the equation of $C_{dl} = i/v$, where C_{dl} was the EDLC, i was the current density and v was the scan rate. Half of the cathodic and anodic current density difference at -2.5 mV (vs. Zn) was plotted vs. scan rate, and linear fit was carried out to obtain

C_{dl} . CV of plating/stripping was tested with graphite paper, Zn and SCE as the working, counter and reference electrodes, respectively. The electrochemical measurements were performed on LANHE CT2001A battery cyclers or Biologic VMP3.

Supporting Figures and Table

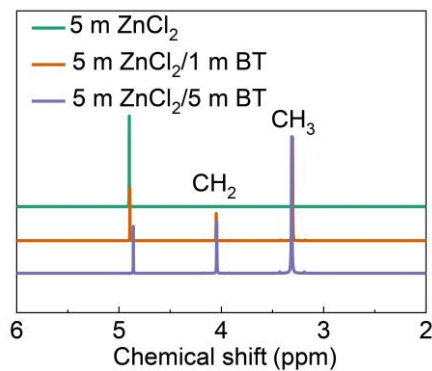


Figure S1. ^1H NMR of the three electrolytes.

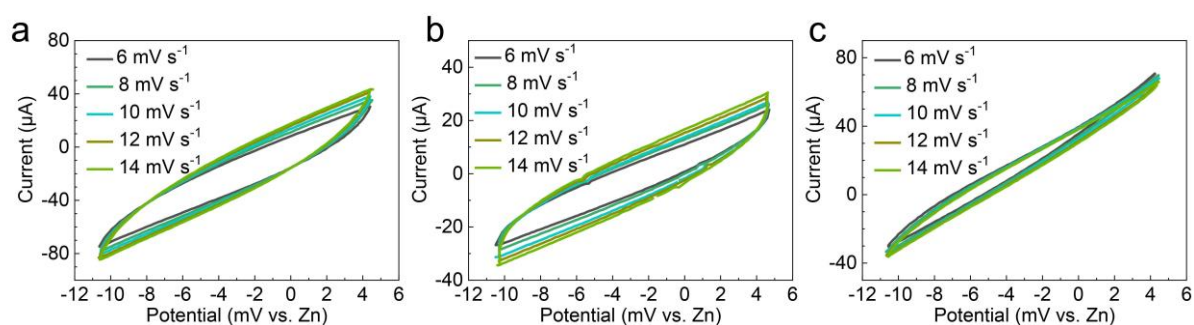


Figure S2. CV curves of Zn electrode in a) 5 m ZnCl_2 , b) 5 m $\text{ZnCl}_2/1$ m BT, c) 5 m $\text{ZnCl}_2/5$ m BT in the non-Faraday region at various scan rates.

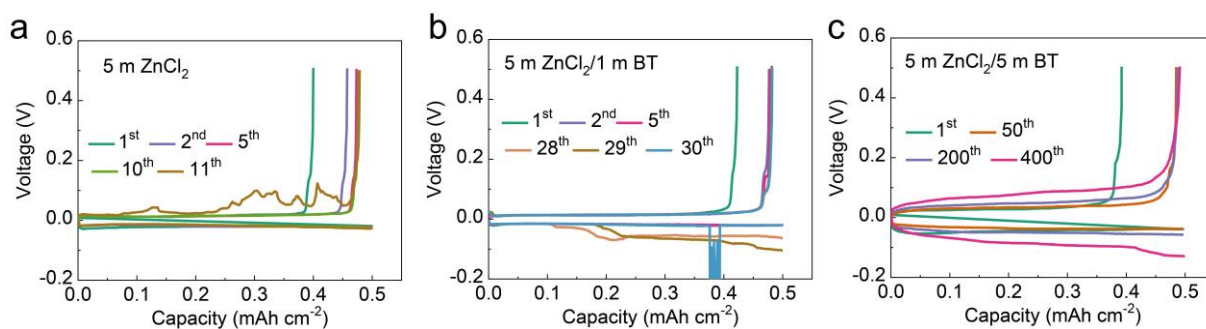


Figure S3. The voltage curves of coulombic efficiency tests in the a) 5 m ZnCl_2 , b) 5 m $\text{ZnCl}_2/1$ m BT, c) 5 m $\text{ZnCl}_2/5$ m BT electrolytes.

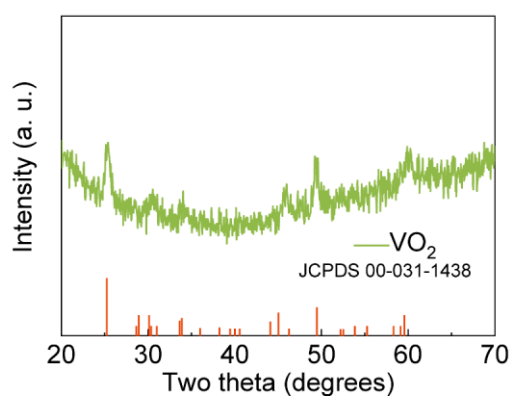


Figure S4. XRD pattern of VO₂.

Table S1. Corrosion parameters of zinc electrode in the three electrolytes.

electrolyte	β_a (mV)	β_c (mV)	E_{corr} (mV)	i_{corr} ($\mu\text{A cm}^{-2}$)
5 m ZnCl ₂	49.0	-49.7	-930	477
5 m ZnCl ₂ /1 m BT	31.0	-31.5	-951	153
5 m ZnCl ₂ /5 m BT	27.5	-30.8	-986	39