

Supporting Information for

## Manipulating Interfacial Stability via Absorption-Competition Mechanism for Long-Lifespan Zn Anode

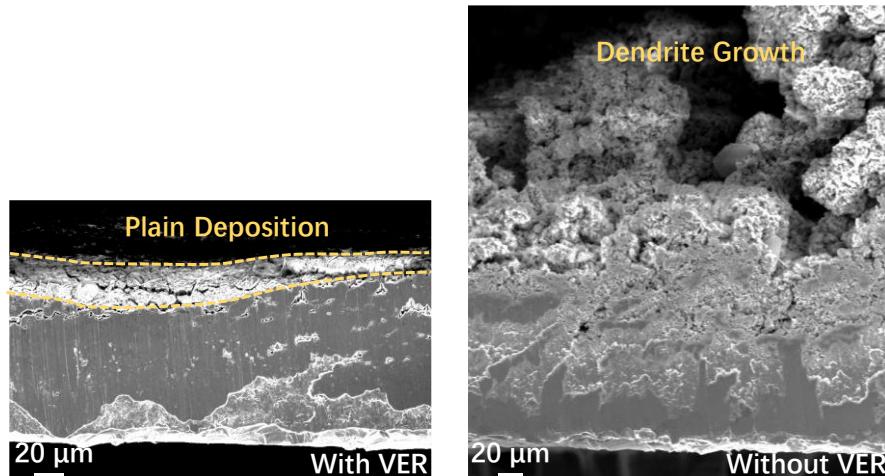
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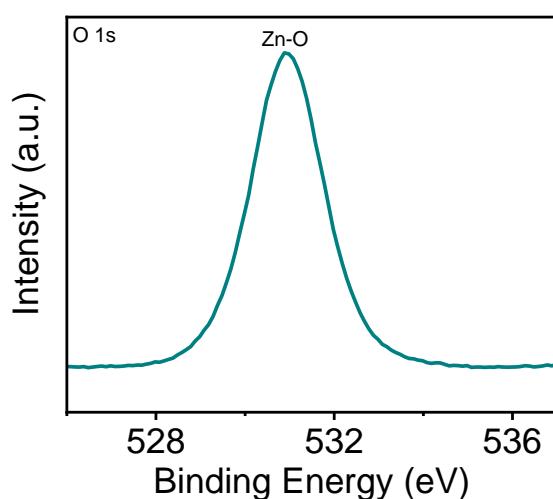
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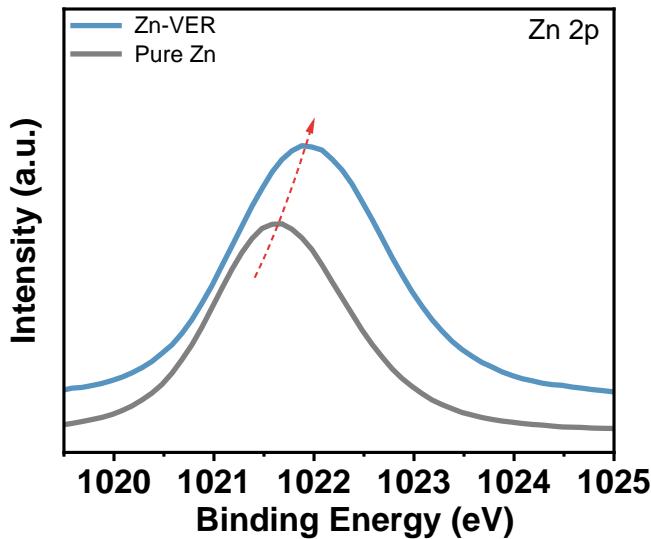
### Supplementary Figures



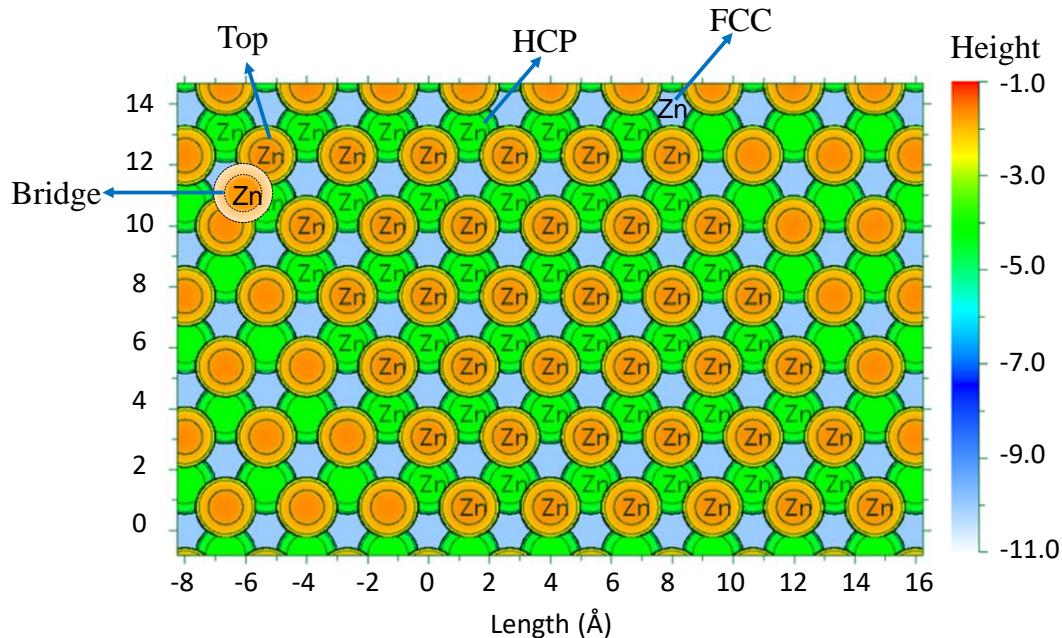
**Fig. S1** Cross-sectional SEM images of Zn sheet surface with (left) and without (right) the veratraldehyde additive



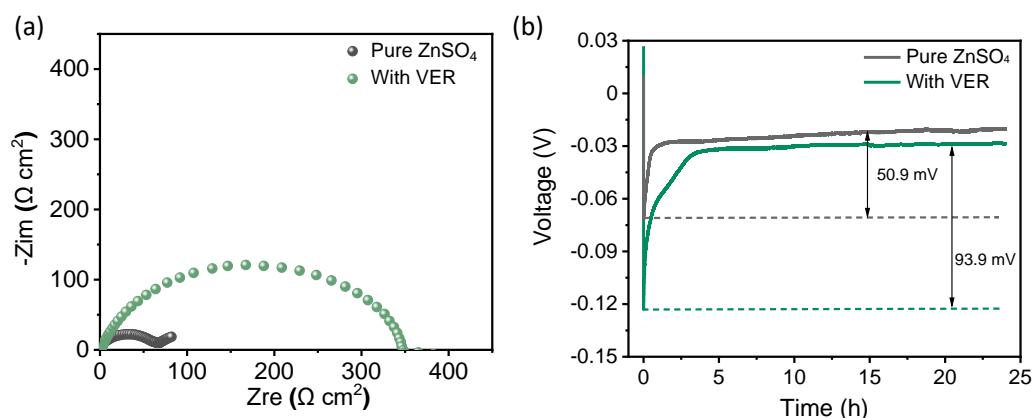
**Fig. S2** The high-resolution XPS spectra of O 1s for the Zn sheet after soaked in DI water



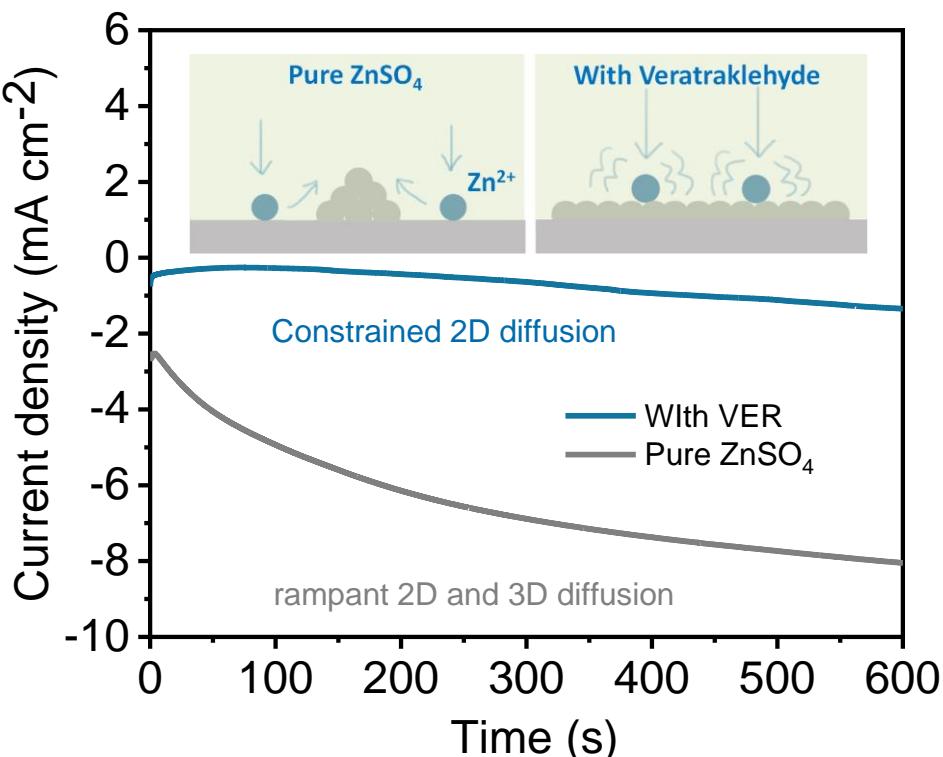
**Fig. S3** Comparison of the high-resolution XPS spectra of Zn 2p between pure Zn sheet and the Zn sheet after soaked in 0.3 g L<sup>-1</sup> veratraldehyde solution



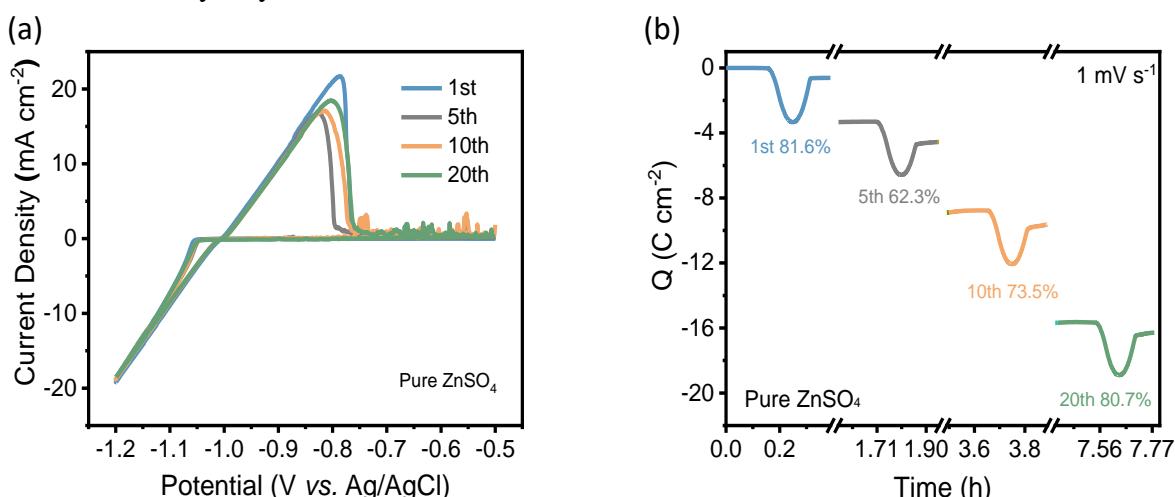
**Fig. S4** Top view of the Zn slab with possible absorbed sites



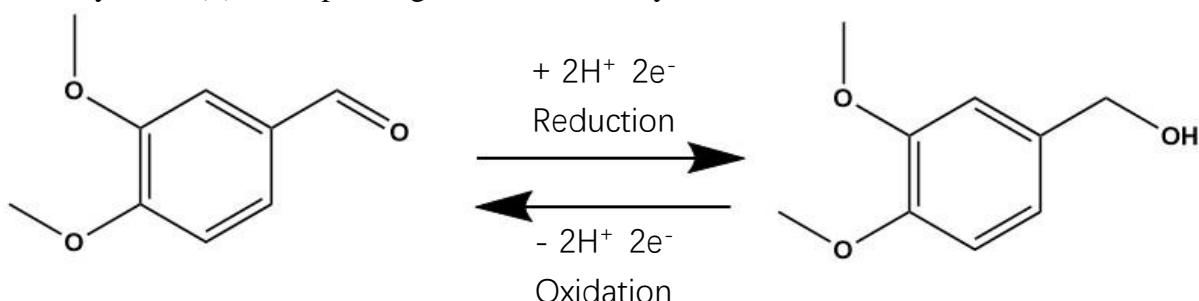
**Fig. S5** (a) EIS and (b) nucleation overpotential comparison of Zn-Zn symmetric cells under electrolyte systems with and without veratraldehyde additive



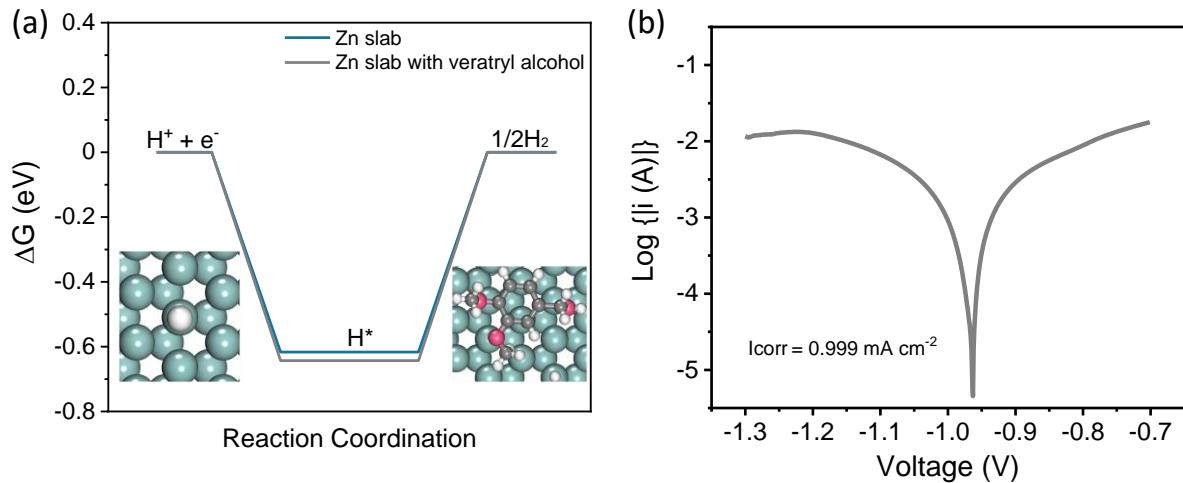
**Fig. S6** Chronoamperometry curves indicating Zn<sup>2+</sup> diffusion process of Zn electrode under different electrolyte system



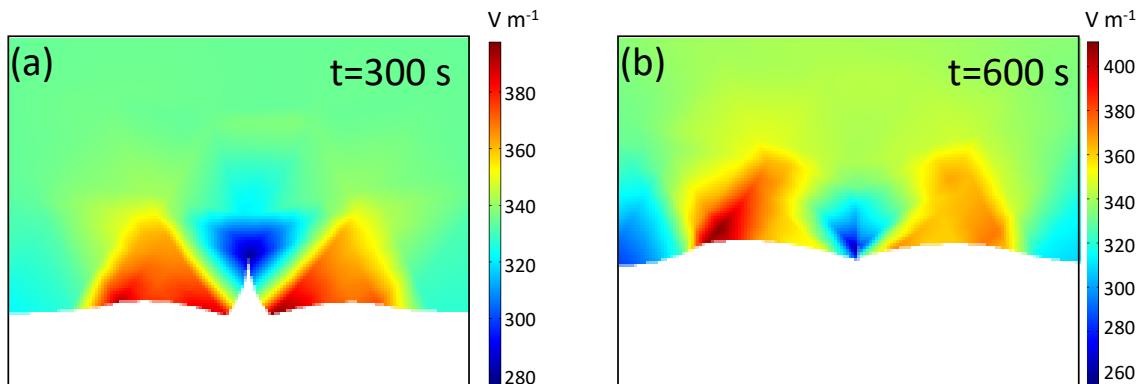
**Fig. S7** (a) CV curves of Zn plating/stripping at a scan rate of 1 mV s<sup>-1</sup> in the pure ZnSO<sub>4</sub> electrolyte and (b) corresponding chronocoulometry curves based on above CV curves



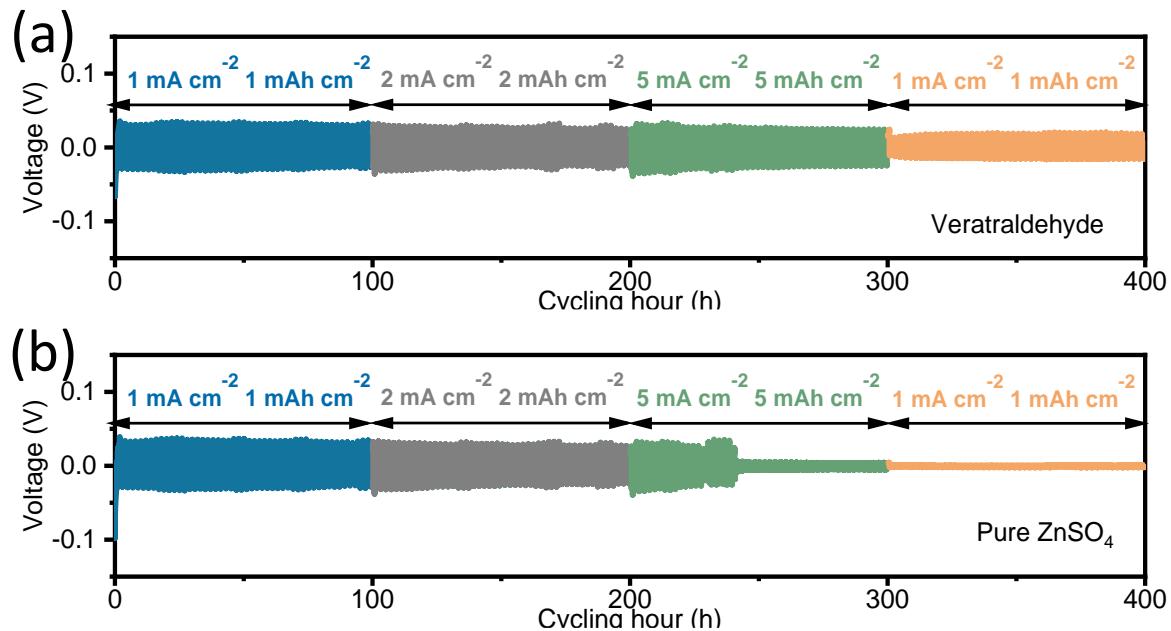
**Fig. S8** Reaction formula of reversible redox process between the veratraldehyde and veratryl alcohol molecules



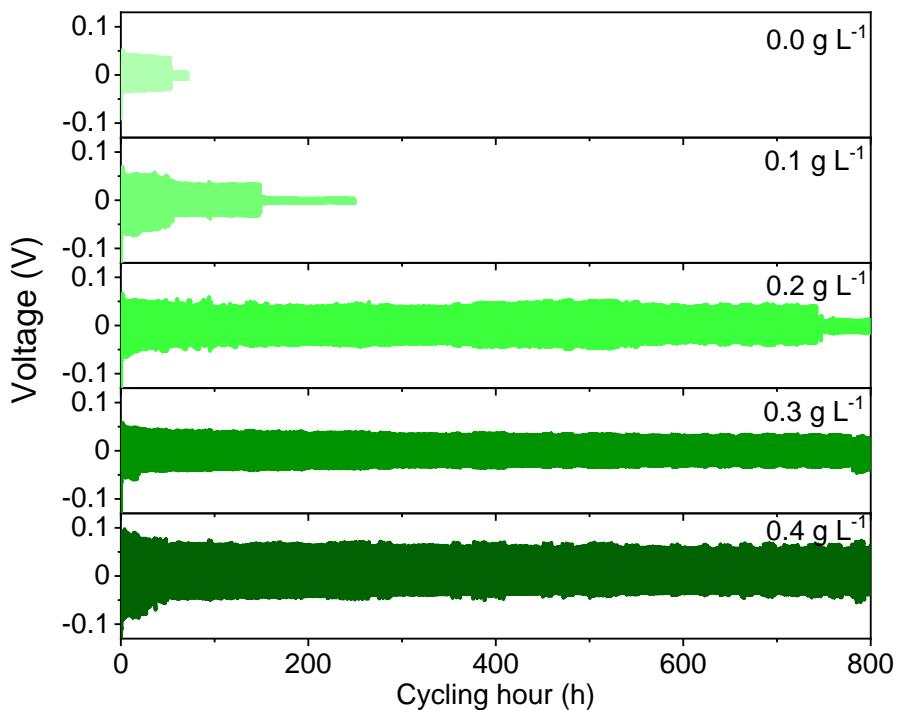
**Fig. S9** (a) The HER diagrams of pure Zn slab and Zn slab with absorbed veratryl alcohol; (b) Tafel plot representing the corrosion behavior under  $\text{ZnSO}_4$ -veratraldehyde electrolyte



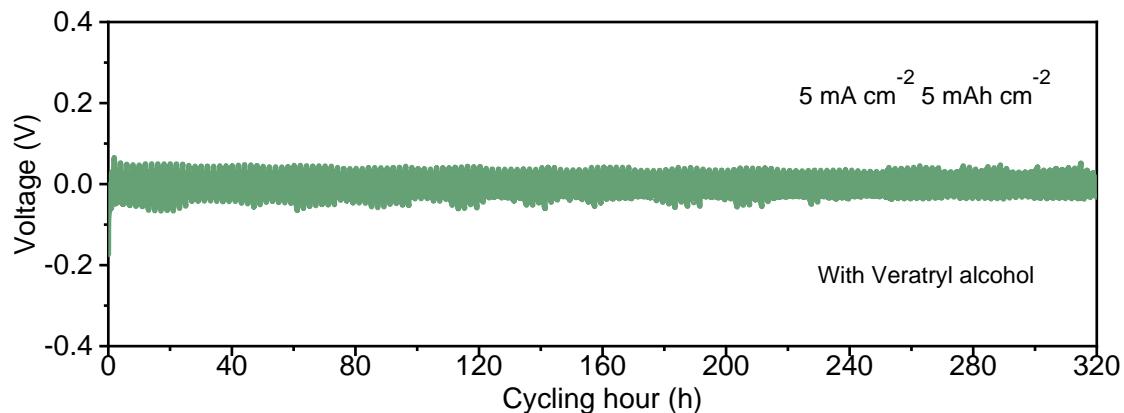
**Fig. S10** Models of electric field intensity distribution under (a)-(b)  $\text{ZnSO}_4$ -veratraldehyde electrolytes at different reaction time



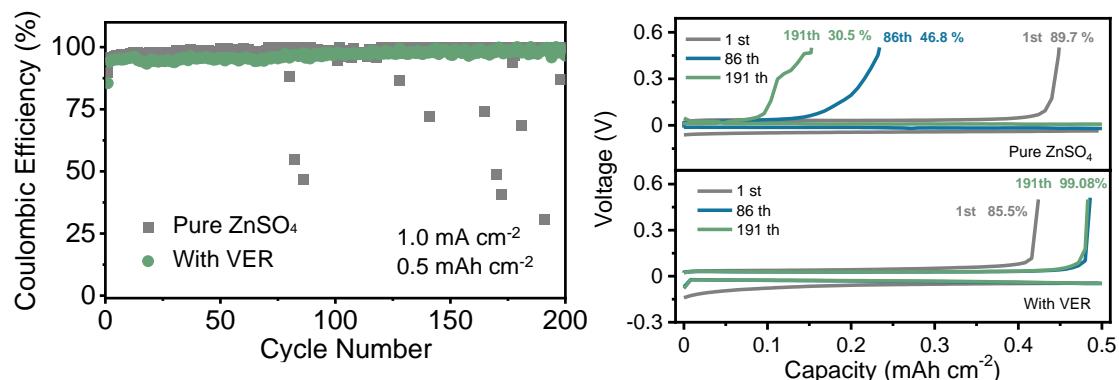
**Fig. S11** Rate cycling performance comparison of Zn-Zn symmetric cells under different current densities and areal capacities of  $1 \text{ mA cm}^{-2}$ ,  $1 \text{ mAh cm}^{-2}$ ;  $2 \text{ mA cm}^{-2}$ ,  $2 \text{ mAh cm}^{-2}$  and  $5 \text{ mA cm}^{-2}$ ,  $5 \text{ mAh cm}^{-2}$  in (a)  $\text{ZnSO}_4$ -veratraldehyde and (b) pure  $\text{ZnSO}_4$  electrolytes



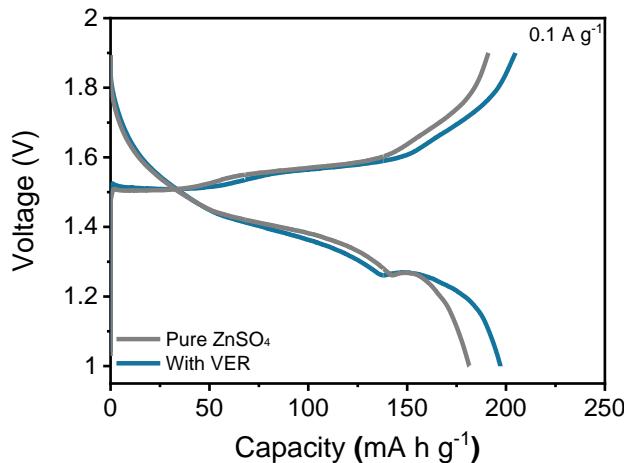
**Fig. S12** Long-term cycling performance comparison of Zn-Zn symmetric cells under current densities and areal capacities of  $5 \text{ mA cm}^{-2}$ ,  $5 \text{ mAh cm}^{-2}$  in  $\text{ZnSO}_4$  electrolytes with different veratraldehyde concentration



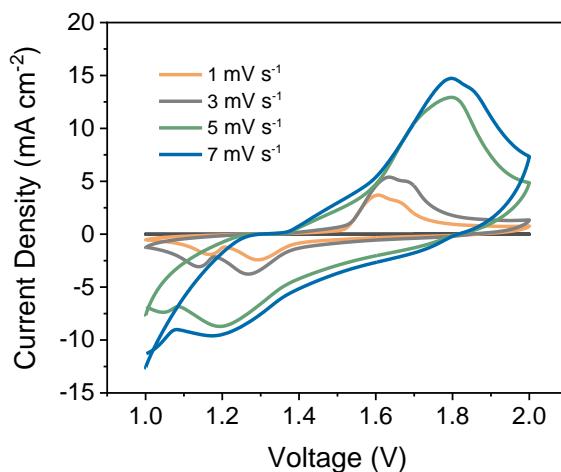
**Fig. S13** Long-term cycling performance comparison of Zn-Zn symmetric cells under current densities and areal capacities of  $5 \text{ mA cm}^{-2}$ ,  $5 \text{ mAh cm}^{-2}$  in  $\text{ZnSO}_4$  electrolytes with alcohol additive



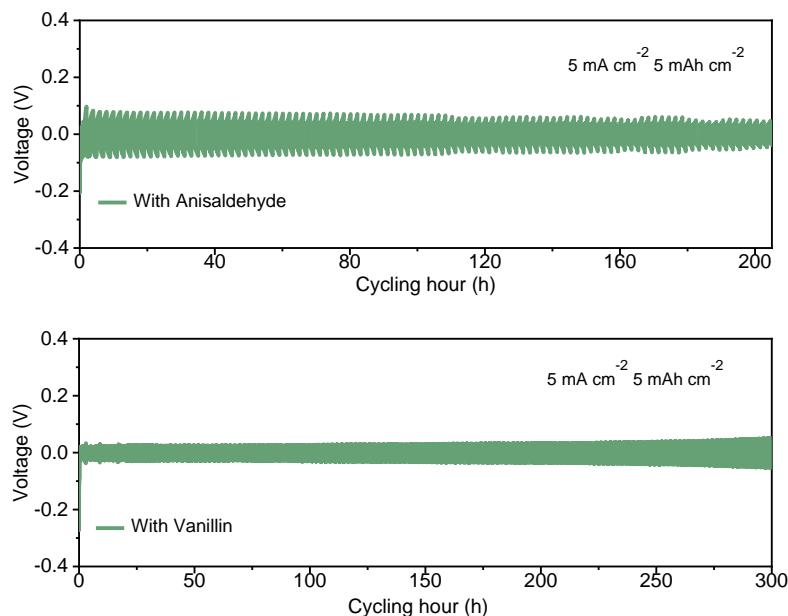
**Fig. S14** Coulombic efficiency (CE) measurements of Zn/Ti cells and corresponding voltage profiles at various cycles under pure  $\text{ZnSO}_4$  and  $\text{ZnSO}_4$ -veratraldehyde electrolytes



**Fig. S15** GCD curves at a current density of  $0.1 \text{ A g}^{-1}$  in different electrolytes



**Fig. S16** CV curves of Zn-MnO<sub>2</sub> batteries under different scan rates ranging from  $1 \text{ mV s}^{-1}$  to  $7 \text{ mV s}^{-1}$



**Fig. S17** Long-term cycling performance comparison of Zn-Zn symmetric cells under current densities and areal capacities of  $5 \text{ mA cm}^{-2}$ ,  $5 \text{ mAh cm}^{-2}$  in ZnSO<sub>4</sub> electrolytes with anisaldehyde and vanillin additives