

Bond-length engineering unlocks ultrahigh-DOD aqueous zinc batteries with relaxed solvation shell

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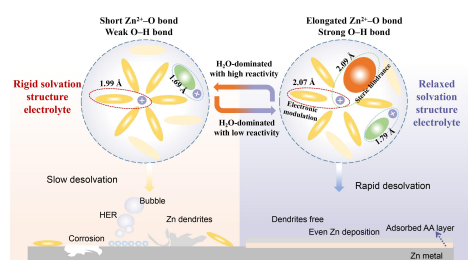
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A bond-length engineering approach strengthens intramolecular O–H bonds and elongates Zn^{2+} – $\text{O}(\text{H}_2\text{O})$ bonds via electronic and steric effects, facilitating Zn^{2+} desolvation and raising the H_2O reduction barrier. This intrinsically suppresses H_2O -induced issues, enabling excellent Zn anode cycling performance even at ultrahigh depth of discharge, current density, and areal capacity.

Bond-length engineering unlocks ultrahigh-DOD aqueous zinc batteries with relaxed solvation shell

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ABSTRACT: Aqueous Zn-ion battery development is challenged by H₂O-induced side reactions and dendrite growth. Interface protective engineering and electrolyte additive including high-concentration salts or organic co-solvents can reduce H₂O molecules, but often at the expense of cost, ionic conductivity, and safety. An approach that mitigates H₂O damage without compromising the merits of aqueous electrolytes is highly desired. Herein, we propose an intrinsic tuning strategy to strengthen the intramolecular O–H bond and elongate the Zn²⁺–O (H₂O) bond length by utilizing electronic and steric hindrance effects, thereby inherently reducing the H₂O reactivity and constructing a relaxed solvation shell. Using ascorbic acid (AA) as a proof-of-concept additive, this bond-length engineering is verified to facilitate Zn²⁺ desolvation and raise the H₂O reduction barrier, leading to even Zn electrodeposition together with inhibition of detrimental side reactions. Therefore, the Zn anode achieves stable cycling for over 380 h under ultrahigh depth of discharge (DOD) of 86.1% together with high current density and areal capacity (100 mA/mAh cm⁻²). Full Zn/NaV₃O₈·1.5H₂O cell also presents remarkably enhanced cycling stability, demonstrating the practicality of this approach for high-performance AZIB.

KEYWORDS: aqueous Zn batteries, Zn anode, bond-length, steric hindrance effect

1 Introduction

The advancement of inherently non-hazardous and low-cost energy storage technologies is essential to the integration of intermittent and fluctuating renewable energy sources into smart grids, particularly in the context of energy storage on a large scale[1-4]. Rechargeable aqueous zinc-ion batteries (AZIBs) have been recognized as a competitive choice in this field, owing to their environmental benignity, natural abundance, low cost, high ionic conductivity and compelling electrochemical performance[5-7]. However, AZIBs are still plagued by the thermodynamically unstable Zn/electrolyte interface, which includes side reactions like HER and dendrite growth, severely impairing the reversibility associated with Zn²⁺/Zn⁰[8, 9]. Specifically, Zn²⁺ ions are typically coordinated by H₂O molecules to form an abundantly water-coordinated [Zn(H₂O)₆]²⁺ structure existing in a dilute benchmark electrolyte. The solvated H₂O molecules with high reactivity in this configuration and other free H₂O trigger detrimental HER and corrosion during Zn deposition process[10, 11]. These undesirable processes induce electric field inhomogeneity across the Zn metal anode. This cause a disordered Zn²⁺ migration together with an inhomogeneous Zn plating pattern, which eventually causes the battery to fail[12, 13]. To mitigate these issues, research efforts have been predominantly directed toward constructing artificial protective layers on the Zn surface to isolate H₂O molecules[14-16], or designing new classes of electrolytes to reduce the number of H₂O molecules, including but not limited to high-concentration electrolytes (HCEs) [17, 18], localized high-concentration electrolytes (LHCEs) [19-21], or weakly solvating electrolytes (WSEs) [22, 23]. Despite the inspiring achievements, these strategies inevitably introduce certain trade-offs. For instance, interfacial modifications often impede ion transport, while introducing high-concentration salts or organic additives in electrolytes may suffer from high cost and elevate viscosity for the former, and low ionic conductivity along with increased flammability for the latter. Therefore, developing optimization methods to effectively alleviate the detrimental effects of H₂O molecules, while retaining the inherent merits of aqueous electrolytes is crucial for achieving stable Zn anodes in AZIBs.

The key to resolve the above issues is to directly decrease the innate reactivity of H₂O molecules by tuning their electronic structure. The reactivity of H₂O molecules is intrinsically linked to the electron density distribution within the molecule and

the strength of its chemical bonds. In the conventional Zn^{2+} solvation structure, coordinated H_2O molecules are polarized by the strong electric field of the Zn^{2+} ion, thus exhibiting high reactivity[24, 25]. Specifically, the high positive charge and charge density of Zn^{2+} enable it to strongly interact with the oxygen atom in H_2O molecules. Such an interaction induces a back-donation of electron from the O–H bonding orbitals towards the oxygen atom and subsequently to the central Zn^{2+} ion. This transfer shifts the electron cloud and weakens the O–H bond strength, facilitating its cleavage. From the perspective of molecular orbital theory, such electron transfer lowers the energy of Lowest Unoccupied Molecular Orbital (LUMO) of the coordinated H_2O molecule, effectively reducing the thermodynamic driving force and the kinetic barrier for the reduction of H_2O molecules at the Zn anode interface, which is favorable for promoting the occurrence of the HER ($2\text{H}_2\text{O} + 2\text{e}^- \rightarrow \text{H}_2 + 2\text{OH}^-$)[26, 27]. Consequently, the strength of the Zn^{2+} –O (H_2O) coordinate bond directly governs the state of O–H bond and the reactivity of the H_2O molecule. Elongating the Zn^{2+} –O bond is equivalent to reducing the bond order of this coordinate interaction. This not only attenuates the polarizing power of Zn^{2+} on the H_2O molecule, but more importantly, diminishes the electron transfer from H_2O to Zn^{2+} . In other words, elongating the Zn^{2+} –O (H_2O) bond length effectively reduces the reactivity of H_2O molecules, raising the energy barrier for HER, thereby essentially suppressing the side reactions.

In this work, we validate the proposed strategy of elongating the Zn^{2+} –O (H_2O) bond length for enhancing Zn anode stability by selecting ascorbic acid (AA) as a proof-of-concept additive. AA was selected due to its ability to effectively elongate the Zn^{2+} –O (H_2O) bond through the combination of electronic and steric hindrance effects, lowering the H_2O molecules reactivity and forming a relaxed solvation structure, thus achieving dendrites- and HER-free Zn anode (Figure 1). Specifically, the efficacy of AA stems from two key criteria for this purpose: i) its carbonyl group has strong coordination capability with Zn^{2+} , enabling competitive entry into the primary solvation sheath (PSS), which increases electron density transfer to Zn^{2+} and reduces electrostatic attraction to coordinated H_2O , thereby elongating Zn^{2+} –O bonds; and ii) its extended conjugated system and multiple polar groups generate steric hindrance, which further expands the typical Zn^{2+} –O bond distance as H_2O species are ejected from the immediate solvation environment, thus yielding a more relaxed arrangement of the solvation shell. The synergetic electronic and steric hindrance effects not only elevate the reduction barrier of H_2O , suppressing HER and corrosion, but also lowers the Zn^{2+} desolvation barrier, promoting uniform Zn deposition. Additionally, the hydroxyl and carbonyl groups of AA can also disrupt the H-bonding network of free H_2O and facilitate preferential adsorption onto the Zn surface, particularly at high-energy “hot spots”, forming a protective layer that further mitigates H_2O -induced detrimental reactions. In the AA-modified electrolyte (ZT-AA), the Zn anode delivers stable operation for >380 h, even when subjected to an ultra-high depth of discharge (DOD, 91.1%) combined with ultrahigh current density and areal capacity ($100 \text{ mA cm}^{-2}/100 \text{ mAh cm}^{-2}$). The ZT cell, by contrast, lasts less than 25 h. The effectiveness of this bond-elongation engineering is further confirmed by the improved cycling stability of full Zn/ $\text{NaV}_3\text{O}_8 \cdot 1.5\text{H}_2\text{O}$ (NVO) cells.

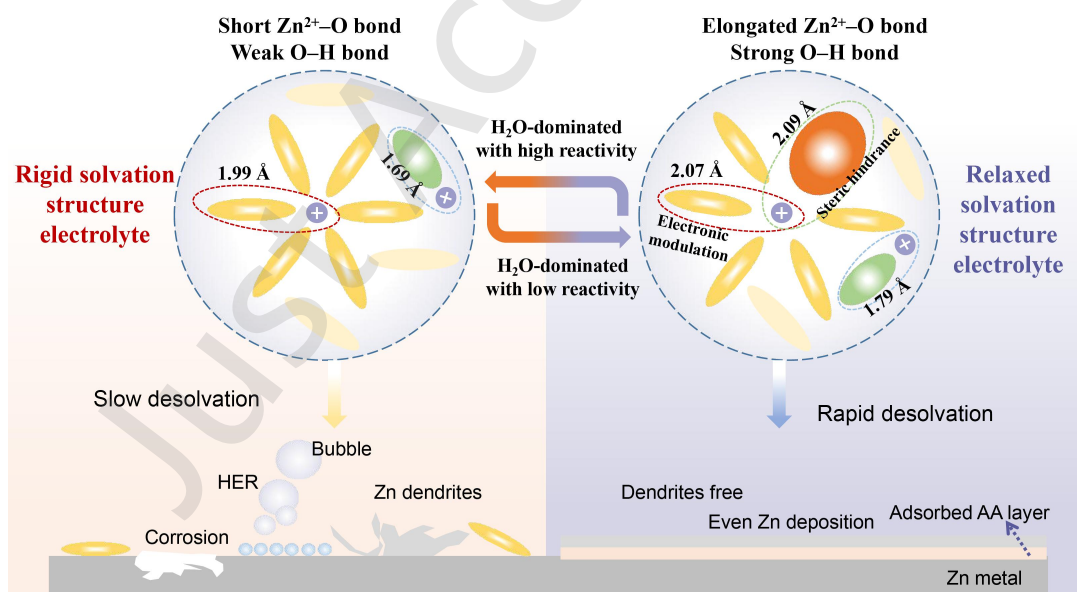


Figure 1. Mechanism schematic diagram for the realization of stable Zn anode through bond-length engineering.

2 Experimental

2.1 Electrolytes preparation

Analytical reagent grade chemicals were used throughout, and none of them were further purified. The ZT electrolyte was produced by putting $\text{Zn}(\text{OTf})_2$ at 2 M in deionized water to yield transparent solution. For the ZT-AA electrolyte, 0.1 M ascorbic acid (AA) was dissolved in 2 M $\text{Zn}(\text{OTf})_2$ electrolyte. To optimize the AA concentration, electrolytes containing 0.05 M and 0.2 M AA in 2 M $\text{Zn}(\text{OTf})_2$ were also prepared and denoted as ZT-005AA and ZT-02AA, respectively, for comparative studies.

2.2 Cathode synthesis

Preparation of NVO material: At 30 °C, the mixture was stirred for 96 h. The product obtained was then given multiple washes with deionized water and subsequently desiccated in a vacuum chamber set to 60 °C for 12 h following the reaction, producing the $\text{NaV}_3\text{O}_8 \cdot 1.5\text{H}_2\text{O}$ (denoted as NVO) sample.

Preparation of NVO electrode: The NVO cathode was prepared by mixing NVO powder, acetylene black, together with polyvinylidene fluoride (PVDF) at a weight proportion of 7:2:1 with an appropriate amount of 1-methyl-2-pyrrolidinone (NMP) solvent. The mixture was kept stirring at room temperature for a period of 4 h to produce a well-mixed slurry. We then coated the resulting slurry onto carbon paper in a uniform manner and dried it in a vacuum oven set to 60 °C for 12 h. The obtained cathode possessed an active material mass loading of approximately 2.75 mg cm^{-2} and 3.30 mg cm^{-2} .

2.3 Cells fabrication

Zn/Cu and Zn/Zn cell: The Zn/Cu and Zn/Zn coin cells were constructed using Cu and Zn foil electrodes, glass fiber separator and 80 μL of ZT or ZT-AA electrolytes. The area and thickness of Cu are 1.13 cm^2 and 0.1 mm, respectively. The area and thickness of Zn foil are 1.13 cm^2 and 0.1 mm or 0.2 mm, respectively. The Zn/NVO coin cells were fabricated with NVO cathode, Zn anode, ZT and ZT-AA electrolytes.

2.4 Pouch cell assembly

The NVO cathodes were fabricated following the same procedure described above, with an active material loading of $\sim 10 \text{ mg cm}^{-2}$ on carbon paper substrates (each electrode area: $4.5 \text{ cm} \times 10 \text{ cm}$). The pouch cell was constructed by alternately stacking four NVO cathodes and five Zn (0.02 mm) anodes, separated by glass fiber separators. The electrolyte was added with a lean electrolyte-to-capacity ratio (E/C) of 15 mL Ah^{-1} and a low negative-to-positive capacity ratio (N/P) of ~ 3.01 . Galvanostatic charge-discharge tests of the pouch cells were conducted at ambient temperature using a battery testing system.

2.5 Characterizations and electrochemical measurements

Materials Characterization: A JSM-6330F field-emission scanning electron microscope (FE-SEM) was employed to characterize the microstructure of the NVO sample. X-ray diffractometry (XRD, D8 ADVANCE) was used to determine the chemical composition of the material. The coordination environment of Zn^{2+} species in ZT and ZT-AA electrolytes was investigated by Raman spectroscopy (Renishaw inVia, 532 nm), Fourier-transform infrared spectroscopy (FTIR) with Vertex70Hyperion3000 instrument, and nuclear magnetic resonance spectroscopy (NMR) performed on a Varian INOVA500NB system (field frequency lock), with the sample temperature held at 25 °C. FE-SEM (JSM-6330F) was used to examine the morphology of Zn anodes after cycling, while XRD (D8 ADVANCE) was employed to determine their chemical composition. The pH values of the electrolytes were measured using a pH meter, and ionic conductivities were determined with a conductivity meter at room temperature. Ultraviolet-Visible Spectroscopy (UV-Vis) absorption spectra of fresh and cycled electrolytes were recorded on a UV-vis spectrophotometer to evaluate the chemical stability of AA.

Electrochemical measurements: The cycling durability, high-rate performance, and Coulombic efficiency of symmetric coin cells using Zn electrodes on both sides and asymmetric coin cells using Zn and Cu electrodes (2032-type) were evaluated using a battery testing system (NEWARE BTS-CT-4008T). In situ optical microscopy coupled with electrochemical measurements monitored Zn plating/stripping behaviors on Zn foils in symmetric cells containing ZT or ZT-AA electrolytes. A CHI 760E electrochemical workstation was employed with a three-electrode cell (working: Zn foil, counter: graphite, reference: SCE) to evaluate corrosion resistance, hydrogen evolution activity, and chronoamperograms (CA) To examine how full Zn/NVO cells behave electrochemically in different electrolytes, we employed cyclic voltammetry (CV), galvanostatic charge/discharge (GCD), and electrochemical impedance spectroscopy (EIS). Importantly, all tests were run on the identical electrochemical workstation.

Computational Details:

Capacity calculations: The specific capacities (C , mAh g^{-1}) of Zn/NVO full cells were calculated from discharge curves in GCD measurements based on the following equations:

$$C = \frac{\int_0^{\Delta t} I \times dt}{m} \quad (1)$$

in which the specific discharge current is denoted by I (mA cm^{-2}), the discharge duration by Δt (h), and the total mass of electrode active material by m (mg).

Activation energy: The activation energies (E_a) were determined from temperature-dependent EIS of Zn/Zn symmetric cells via the Arrhenius relationship:

$$\ln(R_{ct}) = \ln A + \frac{E_a}{RT} \quad (2)$$

Here, R_{ct} (Ω) refers to the resistance at the interface, A is the pre-exponential (frequency) factor, R represents the universal gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$), and T is the temperature in kelvin (absolute temperature).

Ion transference number: Zn^{2+} transference number ($t_{\text{Zn}^{2+}}$) were evaluated using combined chronoamperometry and EIS spectra of Zn cells before and following polarization of Zn cells by the following equation:

$$t_{\text{Zn}^{2+}} = \frac{I_s(\Delta V - I_0 R_0)}{I_0(\Delta V - I_s R_s)} \quad (3)$$

where ΔV (V) is the applied voltage, I_0 (A) and I_s (A) refer to the currents at the start and after reaching steady state, and R_0 (Ω) and R_s (Ω) refer to the interfacial resistance at those two respective times.

The data and calculation process related to the DOD are acquired from the followed equations:

$$DOD(\%) = \frac{C_{actual}}{C_{theoretical}} \times 100\% = \frac{I \times t \times S}{m \times C_{theoretical}} \times 100\% \quad (4)$$

Where C_{actual} (mAh g⁻¹) is actual specific capacity of deposited/stripped Zn, $C_{theoretical}$ (mAh g⁻¹) is theoretical specific capacity of Zn anode (~820 mAh g⁻¹), I (mA cm⁻²) is the current density during plating/stripping process, t (h) is the deposited/stripped time of Zn, S (cm²) and m (g) are actual area and mass of Zn anode, respectively. The value of zinc density is 7.14 g cm⁻³. Specifically,

$$m = 7.14 \text{ g cm}^{-3} \times 0.02 \text{ cm} \times 1.13 \text{ cm}^2 \approx 0.16 \text{ g}$$

$$DOD_{(4.3\%)} = \frac{5 \text{ mA cm}^{-2} \times 1 \text{ h} \times 1.13 \text{ cm}^2}{0.16 \text{ g} \times 820 \text{ mAh g}^{-1}} \times 100\% \approx 4.3\%$$

$$DOD_{(86.1\%)} = \frac{100 \text{ mA cm}^{-2} \times 1 \text{ h} \times 1.13 \text{ cm}^2}{0.16 \text{ g} \times 820 \text{ mAh g}^{-1}} \times 100\% \approx 86.1\%$$

Arithmetic average CE (ACE) from Figure 5a and Figure S19:

$$ACE = \frac{1}{n} \sum_{i=1}^n CE_i \quad (5)$$

Where CE_i is the CE of the i -th cycle, and n is the total number of cycles. The ACE for three parallel cells in ZT are 91.18%, 90.98% and 92.14%, while three parallel cells in ZT-AA are 99.95%, 99.02% and 99.26%.

The standard deviation (S) is obtained from the following equation:

$$S = \sqrt{\frac{\sum (ACE - \overline{CE})^2}{N-1}} \quad (6)$$

Where $\overline{CE}$ is average value of ACE for Zn/Cu cells, N is the quantity of Zn/Cu cell.

For ZT:

$$\overline{CE} = (91.18\% + 90.98\% + 92.14\%) / 3 = 91.43\%$$

$$S = \sqrt{\frac{(91.18\% - 91.43\%)^2 + (90.98\% - 91.43\%)^2 + (92.14\% - 91.43\%)^2}{3-1}} = 0.62\%$$

For ZT-AA:

$$\overline{CE} = (99.95\% + 99.02\% + 99.26\%) / 3 = 99.41\%$$

$$S = \sqrt{\frac{(99.95\% - 99.41\%)^2 + (99.02\% - 99.41\%)^2 + (99.26\% - 99.41\%)^2}{3-1}} = 0.48\%$$

Cumulative CE (CCE) from Figure 5a and Figure S19:

$$CCE = \frac{\sum Q_{stripping}}{\sum Q_{plating}} \times 100\% \quad (7)$$

Where $Q_{stripping}$ and $Q_{plating}$ are the total stripping capacity and total plating capacity, respectively.

The average value of CCE for ZT and ZT-AA cells are 91.45% and 99.69%. The CCE for three parallel cells in ZT are 91.20%, 91.00% and 92.14%, while three parallel cells in ZT-AA are 99.96%, 99.86% and 99.26%. And the S for ZT and ZT-AA are 0.61% and 0.38%. Therefore, the arithmetic average and cumulative CE for ZT are 91.43% $\pm$ 0.62% and 91.45% $\pm$ 0.61%, respectively. The arithmetic average and cumulative CE for ZT-AA are 99.41% $\pm$ 0.48% and 99.69% $\pm$ 0.38%, respectively.

Molecular dynamics simulations: To represent the experimental and control groups, a control system consisting of 5556 H₂O molecules and 200 Zn(OTf)₂ units, as well as an experimental system comprising 5556 H₂O molecules, 200 Zn(OTf)₂, and 40 AA molecules, were constructed using Packmol. The COMPASS III force field was selected for its parameterization against first-principles DFT calculations and extensive experimental data, enabling reliable description of both the Zn²⁺-H₂O and Zn²⁺-AA coordination interactions for statistical structural trends. Initially, the structures of both simulation boxes were energy-minimized with "fine" precision based on the COMPASS III forcefield. Subsequently, equilibrium molecular dynamics simulations were performed in the isothermal-isobaric (NPT) ensemble for 1000 ps, using random initial velocities and a time step of 1 fs. This step aimed to achieve a stable liquid system. During this equilibration, the time evolution of total energy, temperature, and volume was monitored to confirm that these quantities reached stable plateaus before trajectory collection. The COMPASS III forcefield, Ewald summation, and Nosé-Hoover thermostat were employed for interatomic interactions, electrostatic calculations, and temperature control, respectively. Finally, the resulting trajectories were analyzed to compute radial distribution functions (RDF), diffusion coefficients, and average coordination numbers. Given that our electrolyte is a small-molecule solution system where molecular diffusion is relatively fast and coordination equilibrium can be reached within a short timescale, the average coordination numbers combined with RDF analysis sufficiently capture the dominant solvation motifs. All molecular dynamics simulations, hydrogen bond analysis, diffusion coefficient calculations, and RDF evaluations were carried out using the open-source software LAMMPS and VMD.

Density functional theory (DFT) calculation: The GGA-PBE functional was employed for all DFT calculations presented in this work. The simulation used a charge state that corresponds to a real situation, and spin-polarized effects were turned off (non-

polarized treatment). A plane-wave basis set with a kinetic energy cutoff of 310 eV was employed, complemented by ultrasoft pseudopotentials and the Koelling–Harmin relativistic treatment. A convergence criterion of 2×10^{-6} eV/atom was adopted for the self-consistent field (SCF) procedure. Structural optimizations were carried out for molecular clusters and individual components. Subsequently, the binding energy (E) that describes the interaction between species was evaluated with the equation shown here:

$$E = E_{ab} - E_a - E_b \quad (8)$$

in which E_{ab} stands for the overall energy of the associated (bound) species, while E_a and E_b refer to the respective energies of the two separate components prior to their association.

3 Results and discussion

Based on the established Lewis acid-base theory, ascorbic acid (AA) was selected as a model electrolyte additive to validate our proposed bond-elongation engineering. The carbonyl oxygen in AA exhibits higher electron density and more spatially concentrated lone-pair electrons compared to the oxygen in H_2O , owing to the neighboring carbon's ability to withdraw electron density via induction, plus the non-localized nature of the π bond. This feature enhances its Lewis basicity, enabling AA to donate electron pairs more effectively to Zn^{2+} and to strongly interact with Zn^{2+} . Consequently, AA partially substitutes H_2O within the Zn^{2+} coordination sphere, thereby diminishing the Coulombic interaction between Zn^{2+} and water, and concurrently lengthening the Zn^{2+} -O (H_2O) distance. Moreover, AA possesses an extended conjugated system and multiple polar groups (such as enol hydroxyl groups and lactone carbonyl groups), leading to a more diffuse electron cloud than that of H_2O . When AA coordinates to Zn^{2+} , the filled molecular orbitals of AA, especially the lone-pair and π electrons on oxygen, overlap with the electron clouds of adjacent coordinated H_2O molecules. According to the Pauli exclusion principle, the exchange repulsion between electrons of the same spin generates a pronounced steric effect. To lower the system energy, the repelled H_2O molecules shift away from Zn^{2+} , resulting in a further increase in the Zn^{2+} -O (H_2O) bond length and forming a relaxed solvation shell (Figure 2a). Above all of these were firstly proved by the density functional theory (DFT) calculations. Figure 2b displays the optimized adsorption configurations of Zn^{2+} with AA and free H_2O molecules. Notably, the adsorption capacity of AA toward Zn^{2+} is stronger than that of H_2O (-15.0 eV vs. -5.4 eV), demonstrating its preferential adsorption on Zn^{2+} . With the purpose of examining how AA impacts the stability of the Zn anode, we used a solution made by dissolving $Zn(OTf)_2$ together with AA, named as ZT-AA. For use as a reference in comparisons, a formulation of $Zn(OTf)_2$ free of AA was also produced and coded as ZT.

To understand how the AA additive affects the Zn^{2+} solvation structure, Raman spectroscopy, Fourier transform infrared spectroscopy (FTIR) together with ^{67}Zn nuclear magnetic resonance (NMR) measurements were systematically conducted. The peak at 1033.5 cm^{-1} in the Raman spectra represents the OTf^- vibration peak with no obvious shift after introducing AA, indicating that AA cannot enable more OTf^- molecules to participate in the Zn^{2+} solvation structure (Figure S1). Such a result is further supported by the observation that the $-CF_3$ signal for free OTf^- remained at 1027.4 cm^{-1} without shifting in Figure S2a. Similar phenomenons are observed in the vibration peaks of $-CF_3$ and $-SO_3$ at $\sim 1176.4\text{ cm}^{-1}$ and $\sim 1289.4\text{ cm}^{-1}$ respectively (without obvious shifts in both electrolytes), demonstrating the negligible changes of OTf^- in the solvation structure of Zn^{2+} ions (Figure S2b) [28]. It is striking that the peak representing Zn^{2+} in ^{67}Zn NMR spectra shows a significant positive shift upon adding AA, implying the electron density around Zn^{2+} decreases (Figure 2c) [29]. This could be attributed to the coordination of the stronger Lewis basic carbonyl oxygen of AA to Zn^{2+} , which polarizes the bonding electron cloud and induces a rearrangement of the inner-shell electrons of Zn^{2+} through the synergistic effects of enhanced covalent character of the bond and the electron-delocalizing ability of the AA molecule, ultimately leading to a drop in the local charge density surrounding the Zn atomic core. The specific Zn^{2+} solvation structure in the ZT and ZT-AA electrolyte systems were further investigated by Molecular dynamics (MD) simulations. For conventional ZT electrolyte, Zn^{2+} ions are primarily solvated by H_2O molecules in the PSS, which can be attributed to the strong solvation ability of H_2O (Figure S3). Radial distribution function (RDF) and coordination number (CN) analyses further confirm this finding. As shown in Figure S4, a pronounced peak corresponding to Zn^{2+} -O (H_2O) coordination is observed at approximately $\sim 2\text{ \AA}$. In contrast, the peak associated with Zn^{2+} - OTf^- coordination exhibits much weaker intensity. The average coordination numbers of H_2O and OTf^- anions around Zn^{2+} are approximately 5.66 and 0.34, respectively, leading to a predominant solvation structure formulated as $Zn^{2+}(H_2O)_{5.66}(OTf^-)_{0.34}$. From Figure 2d, the AA molecule participates in the solvation structure of Zn^{2+} by occupying coordination sites originally held by H_2O molecules in the ZT-AA electrolyte. The detection of two distinct peaks (at about $2\text{ \AA}$) assigned to Zn^{2+} -O from H_2O and from AA also confirms that AA participates in the coordination environment of Zn^{2+} . More specifically, the PSS of Zn^{2+} comprises 4.52 H_2O molecules, 1.13 AA molecules, and 0.35 OTf^- anions, which can be represented by the formula $Zn^{2+}(H_2O)_{4.52}(OTf^-)_{0.35}(AA)_{1.13}$ (Figure 2e and Figure S5). The variational solvation shell of Zn^{2+} ions with reduced H_2O molecules is conducive to alleviate the HER.

The interaction between H_2O and Zn^{2+} within PSS in different electrolytes is studied in Figure 2f. It is observed that the average Zn^{2+} - H_2O bond length in ZT-AA is approximately $2.07\text{ \AA}$, which is longer than that in ZT ($\sim 1.99\text{ \AA}$), indicating a more relaxed PSS around Zn^{2+} in ZT-AA. Further evidence for the modulation of the variation of bond length within Zn^{2+} solvation structure is provided by X-ray absorption spectroscopy. Figure 2g presents a side-by-side comparison of the Zn K-edge X-ray absorption near-edge structure (XANES) spectra acquired from Zn foil, ZnO, ZT, and ZT-AA. The positive shift of the Zn K-edge in the XANES spectra (inset of Figure 2g) suggests a decreased overall electron density around the Zn center, indicating a weakened shielding effect and an altered coordination environment upon AA introduction. Consistent with this finding, the extended X-ray absorption fine structure (EXAFS) spectra display a characteristic peak at $1\sim 2\text{ \AA}$, assigned to the overall Zn-O

coordination in the first solvation shell (Figure 2h). Upon AA addition, a positive shift in this peak position implies an elongation of the average Zn–O bond distance across the entire first coordination shell, which includes contributions from both H₂O and AA oxygen atoms. [30, 31]. This change is ascribed to that the carbonyl oxygen in AA with stronger Lewis base property more effectively donates its electron pair to Zn²⁺, reducing the extent of electron donation from H₂O. Moreover, the more diffuse electron cloud of the AA molecule due to its extended conjugation and multiple polar groups, overlaps with the electron clouds of adjacent coordinating H₂O molecules, which generates Pauli repulsion, leading to a steric hindrance effect. Both factors collectively weaken the electrostatic interaction between Zn²⁺ and its first-shell ligands, resulting a weakened coordination to reduce desolvation barrier.

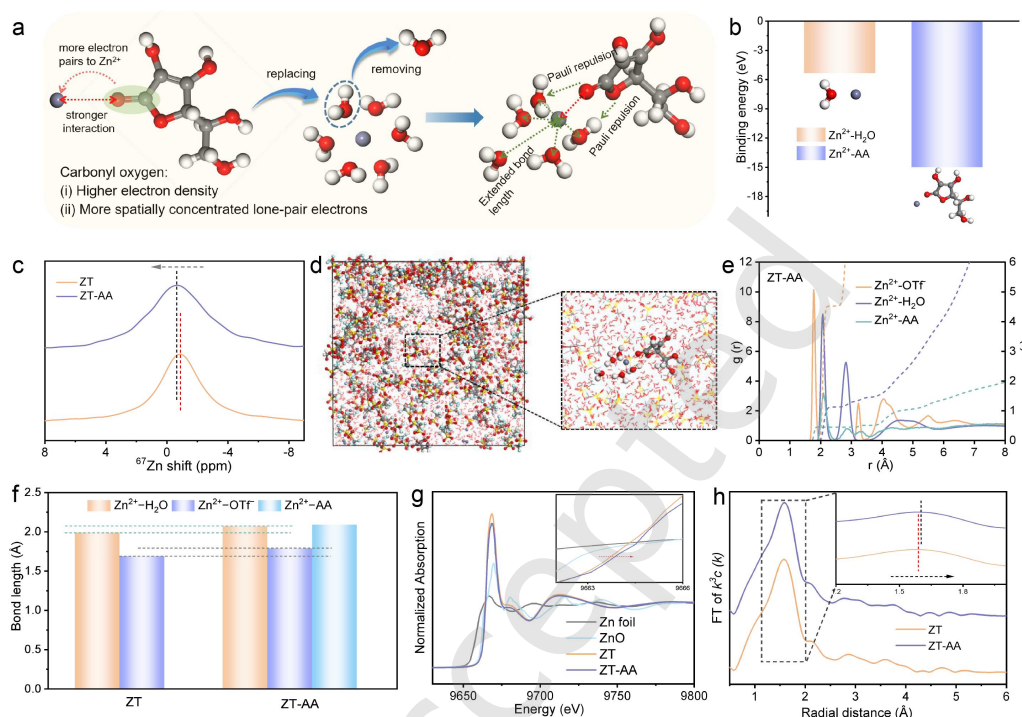


Figure 2. (a) The formation mechanism of the relaxed solvated structure of Zn²⁺ ions. (b) The calculated adsorption energy of Zn²⁺ with H₂O and AA molecules. (c) ⁶⁷Zn NMR spectra of ZT with and without AA. (d) An image of the ZT-AA system from the MD simulation cell. (e) Zn²⁺-O RDFs (H₂O/OTf/AA) for ZT-AA. (f) The average Zn²⁺-H₂O bond length in ZT and ZT-AA. (g) Zn K-edge XANES spectra, (h) EXAFS analysis of the k-space spectra of ZT and ZT-AA electrolytes.

The relaxed solvation structure leads to a lower binding energy for the Zn²⁺(H₂O)_{4.52}(OTf⁻)_{0.35}(AA)_{1.13} complex compared with Zn²⁺(H₂O)_{5.66}(OTf⁻)_{0.34}. The decrease amounts to about 0.95 eV (91.7 kJ mol⁻¹), as shown in Figure 3a, which points to a reduction in the total interaction strength between Zn²⁺ and its coordinated species. The benefits of this reduction include both enhanced Zn²⁺ desolvation kinetics and suppressed water-driven side reactions during Zn plating. Support for these effects comes from electrochemical impedance spectroscopy (EIS) carried out on Zn/Zn symmetric cells with various electrolytes over the temperature interval 0~50 °C (Figure S6). The value of desolvation activation energy (E_a) can be estimated by the Arrhenius relation combining with these present[32, 33]. Figure 3b The E_a for ZT-AA (24.77 kJ mol⁻¹) is reduced compared with the ZT value (26.05 kJ mol⁻¹), confirming the accelerated desolvation kinetics of hydrated Zn²⁺ in ZT-AA, which promote Zn deposition and inhibit H₂O-induced side reactions. The reduced presence of solvated H₂O and the introduction of AA molecules with stronger Lewis basicity can promote the donation of electrons from AA molecules to Zn²⁺. This reduces the strong attraction and polarization effect of the positively charged Zn²⁺ on the electron cloud of H₂O molecules, mitigating the weakening of the O–H bond and the lowering of the LUMO energy level. Additionally, the large conjugated system and multiple polar groups present in AA molecules can elongate the Zn²⁺–O (H₂O) bond distance through steric hindrance effects. This weakens the electron-withdrawing effect of Zn²⁺ on the O–H bond of H₂O molecules, leading to an upward shift in the LUMO energy level, resulting in a decreased thermodynamic tendency for H₂O reduction (Figure 3c). As indicated by the free energy profiles (Figure S7), the computed energy barrier for HER of Zn²⁺(H₂O)_{4.52}(OTf⁻)_{0.35}(AA)_{1.13} complex (–4.89 eV) is higher than that for Zn²⁺(H₂O)_{5.66}(OTf⁻)_{0.34} (–3.81 eV). These results further support that AA contributes dramatically to suppressing the HER, aligning well with the trends discussed above. The reduction reactions were investigated by linear sweep voltammetry (LSV) on Zn/Ti with a scanning speed of 10 mV s⁻¹. The Zn anode in ZT-AA displays lower reduction current density than that in ZT, indicating the effective mitigation of AA for free H₂O-induced HER side reactions (Figure 3d). Additionally, Tafel analysis characterizes the corrosion behavior of Zn anodes in both electrolytes (Figure S8). For ZT-AA, the measured corrosion potential was –0.91 V, which is less negative than the –0.93 V recorded for ZT. Additionally, the corrosion current density was 0.386 mA cm⁻², substantially lower than that of ZT (0.788 mA cm⁻²). These suggest superior corrosion resistance enabled by AA incorporation.

Ascribed to multiple hydroxyl groups (–OH) and carbonyl groups (–C=O), which concurrently possess hydrogen atoms as

donor and oxygen atoms with lone electron pairs serving as hydrogen-bond accepting sites, the AA molecule can competitively integrate into the H-bond network of H₂O molecules to strengthen hydrogen bond network of AA-H₂O. This partially displaces the original hydrogen bonds between H₂O-H₂O, thus decreasing the H-bond number of H₂O in the ZT-AA electrolyte (Figure S9), and disrupts the inherent dynamic tetrahedral network with long-range order in H₂O, transforming it into more localized and short-range H-bonding structures. Such structural change hinders the Grotthuss mechanism that relies on a continuous H-bond network for ultra-fast proton transfer, which effectively slows the kinetics of the HER. This phenomenon was also proved by the ¹H NMR spectra of H₂O from ZT and ZT-AA[34]. As shown in Figure S10, a significant positive shift can be observed in the ¹H chemical shift of H₂O with AA additive. This leads to a reduction in the number of hydrogen bonds compared to the ZT system, resulting in restricted reactivity of free H₂O and mitigation of parasitic reactions. Quantum chemistry simulation results presented in Figure 3e-f reveal a lower minimum and maximum electrostatic potential for the Zn²⁺(H₂O)_{4.52}(OTf⁻)_{0.35}(AA)_{1.13} (0.11 and 0.35 hartree) compared to Zn²⁺(H₂O)_{5.66}(OTf⁻)_{0.34} (0.27 and 0.38 hartree), which demonstrates a weaker attraction of Zn²⁺(H₂O)_{4.52}(OTf⁻)_{0.35}(AA)_{1.13} to anions, favoring enhanced diffusion kinetics of Zn²⁺ ions. The above result was further verified by the higher diffusion coefficients for Zn²⁺ in ZT-AA (5.850×10⁻¹⁰ m² s⁻¹) than that in ZT (3.015×10⁻¹⁰ m² s⁻¹), and a higher transference number (*t*_{Zn²⁺}) in the ZT-AA (0.65) compared to ZT (0.48), as shown in Figure S11 and Figure S12, respectively. Moreover, the stronger adsorption of AA molecules on the Zn surface (−1.18 eV vs −0.98 eV for free H₂O molecules (Figure S13 and Figure S14) suggests that AA can prevent some H₂O molecules from adsorbing at the electrode interface, mitigating electrode corrosion. To probe the electrolyte/electrode interfacial interaction, contact-angle measurements were performed. The results (Figure S15) reveal that the addition of AA substantially enhances the wettability of the Zn surface, as reflected by the reduction in contact angle from 65.8° to 50.2°. Such improved hydrophilicity is conducive to reducing the interfacial barrier and accelerating Zn²⁺ transport, which are further verified by Nyquist plots of Zn/Zn cells in ZT and ZT-AA electrolytes (Figure S16). Stimulated by these distinct merits, the ZT-AA electrolyte ensures faster Zn deposition kinetics, as well as uniform Zn nucleation and deposition. Specifically, exchange current density (*i*₀) analysis evaluates Zn deposition kinetics (Figures 3g and S17). The ZT-AA electrolyte exhibits increased *i*₀ (2.46 mA cm⁻²) in comparison with ZT (1.68 mA cm⁻²), confirming accelerated deposition kinetics. This kinetic enhancement promotes homogeneous deposition of Zn during subsequent growth stages, thereby improving Coulombic efficiency (CE) and cycling stability of Zn anode, especially during operation at a high current density. Chronoamperometry (CA) tests at an overpotential of −150 mV were employed to characterize the Zn²⁺ nucleation and deposition pattern. Figure 3h reveals continuous current density increase in ZT within 300 s, signifying uncontrolled two-dimensional (2D) nucleation and disordered Zn growth. The tendency for deposition to self-accelerate in pursuit of minimal surface energy aggravates both the inhomogeneity of Zn deposition and the formation of dendrites[35, 36]. Conversely, ZT-AA system presents a short period of increased current density, followed by a stable low current density. This shift promotes even Zn plating by enhancing the uniformity of nucleation site and ensuring a smooth electric field distribution. To study the influence of AA concentration on the physicochemical properties of the electrolyte, we prepared 2 M Zn(OTf)₂ electrolytes with varying AA concentrations (0.05 and 0.2 M), denoted as ZT, ZT-005AA and ZT-02AA, respectively, except for 0.1 M AA (ZT-AA or ZT-01AA). The pH and ionic conductivity of these electrolytes were systematically measured in Figure S18. As the AA concentration increased from 0 to 0.2 M, the pH gradually decreased from 4.6 to 3.8, while the ionic conductivity slightly increased from 24.3 to 25.3 mS cm⁻¹. The pH decrease is attributed to the mild acidity of AA. The slight increase in ionic conductivity may result from partial replacement of water molecules by AA, which reduces local viscosity and facilitates ion migration.

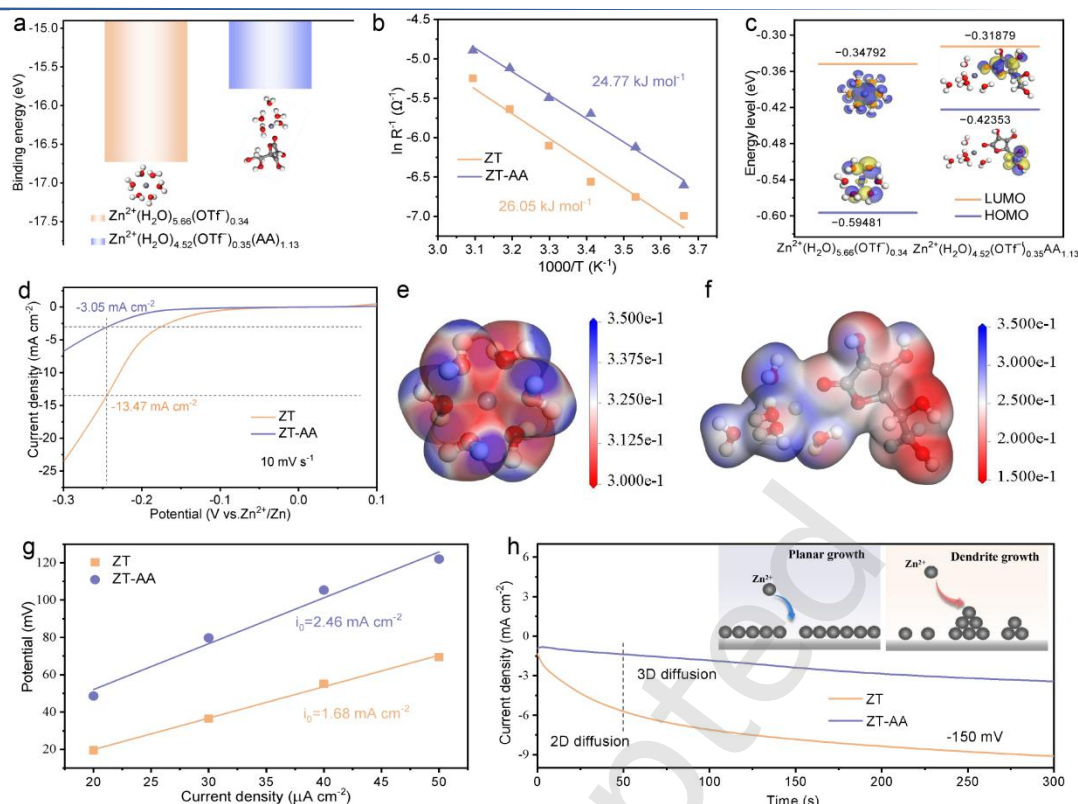


Figure 3. (a) Binding energies of Zn²⁺ and PSS in different electrolytes calculated using DFT. (b) Comparison of E_a of Zn²⁺ desolvation in different electrolytes. (c) Highest Occupied Molecular Orbital (HOMO) and LUMO of Zn²⁺-H₂O in different solvation structure of Zn²⁺. (d) The LSV response of Zn/Ti cells operated with ZT versus ZT-AA electrolytes. Electrostatic potential mapping of (e) Zn²⁺(H₂O)_{5.66}(OTf)_{0.34} and (f) Zn²⁺(H₂O)_{4.52}(OTf)_{0.35}(AA)_{1.13}. (g) Kinetic parameter *i*₀ for Zn reduction in various electrolytes. (h) Comparison of CA traces for Zn/Ti cells employing ZT and ZT-AA as the electrolyte.

To further investigate the effect of AA additive on Zn plating/stripping behavior, scanning electron microscope (SEM) imaging was performed to monitor the morphological changes occurring on the Zn electrodes in different electrolyte systems under 5 mA/mAh cm⁻². As revealed in Figure 4a, the Zn anode surface in the ZT electrolyte develops irregular protrusions and aggregated Zn deposits in random orientations after the initial plating cycle. Following subsequent stripping, incompletely dissolved Zn residues and corrosive areas are observed on the Zn surface in Figure 4b. These morphological degradations progressively exacerbate during the follow-up plating/stripping cycles, ultimately triggering dendrites growth (Figure 4c-e). Notably, Zn anode cycled in ZT-AA maintains compact and uniform morphology throughout plating stages (Figure 4f, h, j). Post-stripping characterization reveals smooth surfaces without any dendrite protrusions or corrosion products (Figure 4g, i), confirming the AA additive can inhibit dendrite formation and mitigate parasitic reactions. This inhibitory influence of AA additive on hydrogen evolution and dendrite growth was also observed through operando optical microscopy images. In the ZT system, uneven deposition and bubbles are detected on the Zn surface as early as 5 min (Figure 4k). With the passage of deposition time, bubbles and dendrites become increasingly evident. Conversely, ZT-AA enables compact and homogeneous Zn deposition devoid of bubbles and dendrite growth even after 30 min, as evidenced in Figure 4l. The mitigation of parasitic reactions on Zn anodes was further confirmed by X-ray diffraction (XRD). Characteristic diffraction peaks ascribed to zinc triflate hydroxide hydrate (Zn_xOTf_y(OH)_{2x-y}·H₂O) are observed on cycled Zn anodes in ZT, whereas these byproducts are absent in ZT-AA (Figure 4m). This highlight further confirms the effective inhibition of corrosion reactions on the Zn anode surface.

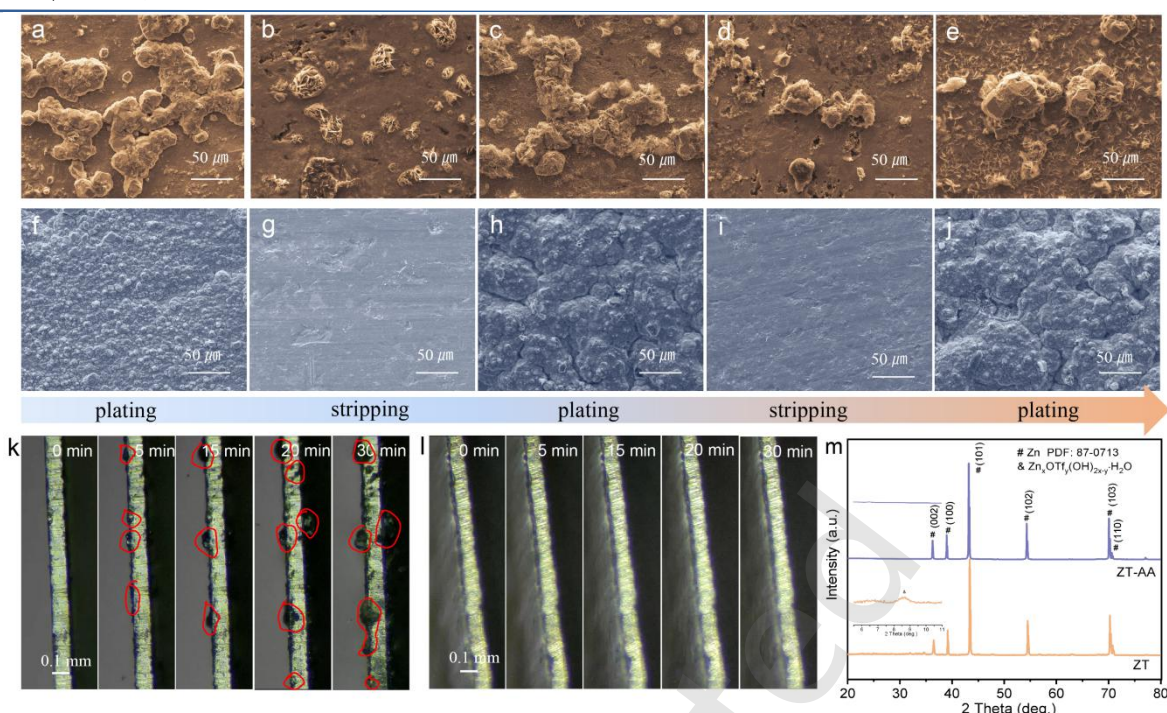


Figure 4. Post-cycling SEM characterization of Zn foil in ZT(a-e) without AA and (f-j) with AA electrolytes. Time-resolved in situ optical microscopy images captured at the Zn/electrolyte boundary while plating and stripping Zn at 5 mA cm^{-2} correspond to (k) ZT and (l) ZT-AA electrolytes. (m) XRD patterns obtained from Zn foil following 50 charge/discharge cycles conducted at $5 \text{ mA}/5 \text{ mAh cm}^{-2}$ in different electrolyte systems.

The cycling reversibility and stability of Zn anodes with ZT and ZT-AA electrolytes were evaluated by asymmetrical Zn/Cu and symmetrical Zn/Zn cells. Coulombic efficiency (CE) as an important parameter, was firstly assessed by testing Zn/Cu cells in two electrolyte systems at the condition of $1 \text{ mA}/\text{mAh cm}^{-2}$. Figure 5a-b reveal that the Zn/Cu asymmetric cells with ZT-AA deliver an exceptionally prolonged cycle life of over 2500 h (1250 cycles). Based on three parallel cells (Figure S19a-b), the arithmetic average and cumulative CE for ZT-AA are $99.41\% \pm 0.48\%$ and $99.69\% \pm 0.38\%$, respectively (detailed calculation process can be found in the Supplementary Information), collectively demonstrating highly reversible Zn deposition with minimal side reactions. In stark contrast, the Zn/Cu cell with the ZT electrolyte fails rapidly within approximately 20 cycles. Combine with other parallel cells (Figure S19c-d), ZT cell deliver $91.43\% \pm 0.62\%$ of arithmetic average CE and $91.45\% \pm 0.61\%$ of cumulative CE. Rate capability was assessed with Zn/Zn symmetric cells that were subjected to cycling across a current density window of $1\sim 10 \text{ mA cm}^{-2}$ at a constant areal capacity of 1 mAh cm^{-2} . The ZT cell abruptly fails to work at 2 mA cm^{-2} (Figure 5c), while the ZT-AA cell sustains stable voltage profiles throughout all the test conditions and recovers its initial voltage profile when the current density returns to the initial current density, demonstrating the enhanced rate performance. Such an enhancement stems from reconfigured Zn^{2+} solvation structure with attenuated reactivity of solvated H_2O , inhibiting dendritic growth and suppressing adverse side reactions.

Subsequently, Zn/Zn symmetric cells employing ZT with/without AA were constructed and then subjected to cycling at $5 \text{ mA}/\text{mAh cm}^{-2}$ with a DOD of 4.3% to evaluate Zn anode stability. As expected, the symmetric cell with ZT-AA owns exceptional cycling stability for over 2000 h without the phenomenon of violent voltage fluctuations or short circuits (Figure 5d and Figure S20). In striking contrast, the ZT-based cell experiences significant voltage fluctuations and rapid failure within merely 34 h, attributable to dendrite formation and parasitic reactions. This pronounced disparity indicates the effectiveness of AA incorporation in enhancing the cycling performance of the Zn anode. For practical aqueous Zn-ion batteries, high DOD is essential to maximize energy density. Thus, the cycling stability of Zn anode was further evaluated at an ultrahigh DOD of 86.1% under extreme conditions with a current density of $100 \text{ mA}/\text{mAh cm}^{-2}$. Employing ZT-AA electrolyte, the symmetric cell retains stable operation for 387 h, and accumulated a plated capacity (CPC) of 38.7 Ah cm^{-2} . Under identical conditions, the ZT-based cell can only be stably operated for approximately 23 h (Figure 5e and Figure S21). This remarkable improvement demonstrates that the AA additive greatly enhances practical application value of AZIBs. Besides, the superior electrochemical characteristics of Zn anode with ZT-AA, including ultrahigh CPC and superior durability even at high current densities, areal capacities and DODs, surpasses the performance of Zn anodes from recently published works, as summarized in Figure 5f, g[22, 25, 31, 37-41]. We conducted SEM on Zn electrodes to characterize their morphology. These electrodes had been cycled (plating/stripping) at 100 mA cm^{-2} and 100 mAh cm^{-2} . The ZT-based Zn anode displays dendritic protrusions and heterogeneous deposits, indicative of localized nucleation that ultimately induces dendrite growth and cell failure (Figure S22). Conversely, the ZT-AA electrolyte yields remarkably compact and uniform Zn deposition without morphological irregularities. This distinct contrast indicates the pivotal role of AA in regulating interfacial chemistry for highly reversible Zn electrodeposition. Subsequently, Zn/Cu asymmetric cells with different AA concentrations were assembled to evaluate the reversibility of Zn plating/stripping in electrolytes with different AA concentrations. As shown in the Figure S23, ZT-005AA

and ZT-02AA cells delivered stable cycling for approximately 512 h and 160 h, respectively, both significantly outperforming the ZT electrolyte, yet neither matched the performance of ZT-AA (~2500 h). Such phenomenon can be attributed to the insufficient solvation-regulating effect at 0.05 M AA, and to the excessively acidic environment at 0.2 M AA, which induces notable chemical corrosion of the Zn anode. This hypothesis is corroborated by SEM. Figure S24 shows the Zn anode cycled in ZT-02AA displayed obvious corrosion pits. To evaluate the chemical stability of AA during prolonged cycling, Ultraviolet-Visible Spectroscopy (UV-Vis) spectroscopy was performed on the ZT-01AA electrolyte before and after cycling. In both cases, a characteristic absorption peak at ~261 nm, corresponding to the conjugated double-bond system of AA, was clearly observed with no significant shift or intensity change (Figure S25). This indicates that AA remains chemically stable under the operating conditions and does not undergo appreciable decomposition or parasitic reactions. Furthermore, to verify the generality of our bond-length engineering strategy beyond AA, we selected two additional organic additives, D-glucose (DG) and aspartic acid (AC), both of which possess strong Zn^{2+} -coordinating groups (e.g., hydroxyl and carbonyl/carboxyl) and extended polar moieties capable of inducing steric hindrance. The cycling performance of Zn/Zn symmetric cells in electrolytes with DG and AC additives (denoted as ZT-DG and ZT-AC, correspondingly) was evaluated at 5 mA cm^{-2} and 5 mAh cm^{-2} . As shown in Figure S26, the ZT-AC and ZT-DG cells achieved stable lifespans of approximately 429 h and 270 h, respectively, markedly surpassing the ZT cell. These results demonstrate the generality of our bond-length engineering strategy to stabilize the Zn anode.

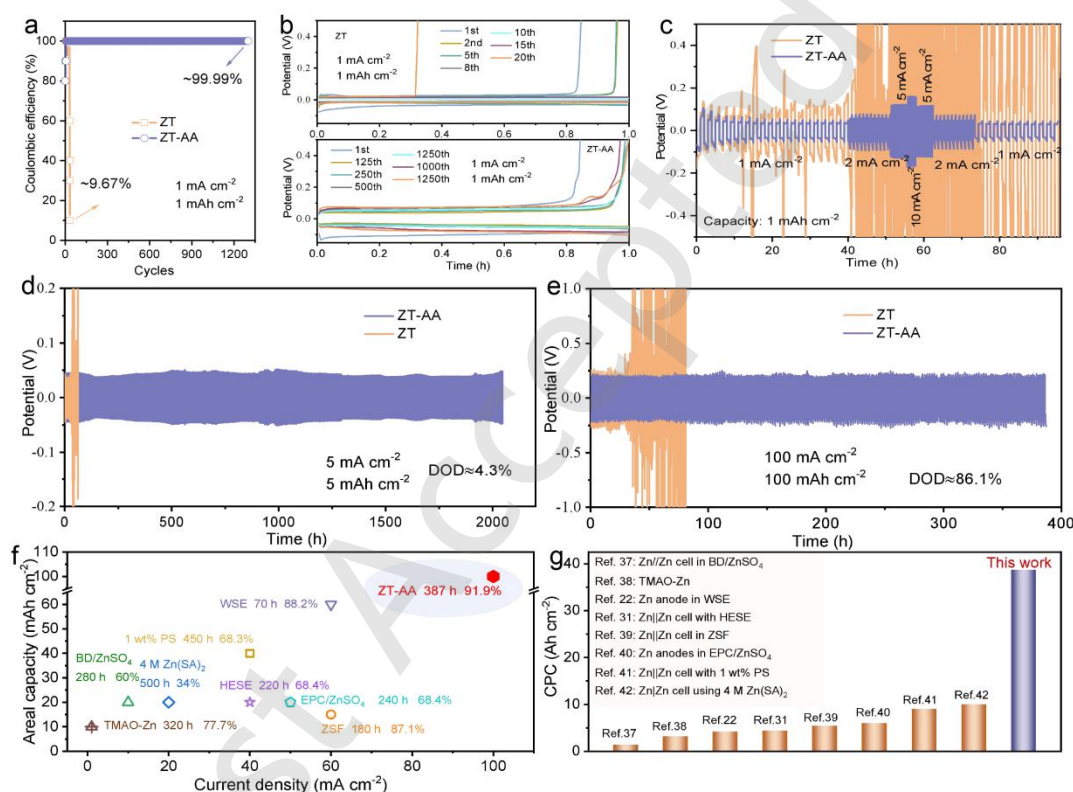


Figure 5. (a) CE recorded for Zn/Cu cells using various electrolyte formulations, tested at 1 mA cm^{-2} and 1 mAh cm^{-2} , and (b) the voltage curves obtained at different cycle numbers. (c) Rate capability comparison of Zn anodes in ZT without/with AA additive. Cycling curves obtained under constant current for symmetric cells with distinct electrolytes at (d) 5 mA cm^{-2} and 5 mAh cm^{-2} , (e) 100 mA cm^{-2} and 100 mAh cm^{-2} . (f) The graph compares the ZT-AA symmetric cell with recently reported Zn-metal anodes stabilized by different approaches across four metrics (current density, areal capacity, cycle life, and DOD). (g) Comparison of CPC of symmetric cells with ZT-AA and that of Zn anodes covered in recently conducted investigations[22, 25, 31, 37-41].

The real-world applicability of ZT-AA electrolyte was demonstrated using a full-cell setup assembled with a $\text{NaV}_3\text{O}_8 \cdot 1.5\text{H}_2\text{O}$ (NVO) cathode and a Zn foil anode (Figure 6a). As illustrated in Figure S27, the XRD pattern of NVO aligns well with $\text{NaV}_3\text{O}_8 \cdot 1.5\text{H}_2\text{O}$ (JCPDS#16-0601), demonstrating its successful synthesis. Furthermore, SEM images in Figure S28 show that the as-synthesized NVO exhibits a belt-shaped nanostructure at the nanoscale. A Zn/NVO cell with ZT-AA electrolyte gives the following cyclic voltammetry (CV) curve shows identical pairs of redox peaks compared to that of ZT electrolyte, indicating that the AA additive introduces no any side reactions to affect the energy storage mechanism of NVO cathode (Figure 6b). Notably, the Zn/NVO cell employing the ZT-AA electrolyte exhibits smaller polarization (253/269 mV) than its counterpart with ZT (301/330 mV). Galvanostatic charge-discharge (GCD) profiles in Figure 6c reveal comparable capacities for both Zn/NVO cells, while ZT-AA exhibits reduced voltage polarization, suggesting enhanced overall reaction kinetics. Figure 6d compares the rate capability of ZT and ZT-AA batteries. At 0.1 A g^{-1} , both devices deliver similar capacities (416.7 mAh g^{-1} for ZT vs. 419.0 mAh g^{-1} for ZT-AA). However, ZT exhibits rapid capacity fading with increasing cycles, and fails completely at the 8th cycle. In contrast, ZT-AA maintains operation up to 5 A g^{-1} with a retained capacity of 159.1 mAh g^{-1} , demonstrating its superior rate performance. After 70 cycles, the current density was switched back to 0.1 A g^{-1} , and the ZT-AA cell's capacity

returned to its full value, thus indicating the excellent cycling reversibility of ZT-AA. EIS curve for the Zn/NVO cell with ZT-AA reveals smaller impedance (R_{ct}) and Warburg resistance (R_w) compared to ZT (Figure S29). Given identical cell components except electrolyte, this diminished resistance is primarily attributed to the regulation of metal-molecule interfacial adsorption by AA, which facilitates enhanced ion transport kinetics and thus enhances rate capability. The cycling performance of both batteries was further analyzed at 2 A g^{-1} . As exhibited in Figure 6e, a quick capacity decay is observed for the ZT-containing cell once it reaches only 30 cycles. Subsequent Zn foil replacement temporarily restores capacity, yet the quick capacity decay recurs after ~ 25 cycles. By comparison, the full cell employing ZT-AA achieves 96.73% capacity retention with 99.41% CE after ~ 1380 cycles (Figure 6f), confirming the effectiveness of AA additive in Zn anode stabilization. The self-discharge response of Zn/NVO cells containing distinct electrolytes was also evaluated in Figure 6g, h. After resting for 24 h, the Zn/NVO cell with ZT-AA kept 79.8% of its initial capacity, whereas the ZT cell only held 64.2%. Such an improvement indicates the effective inhibition of spontaneous parasitic reactions on Zn anode by AA additive. To further evaluate the practical potential of the ZT-AA electrolyte, we assembled Zn/NVO pouch cells with a high active-material loading of $\sim 10 \text{ mg cm}^{-2}$, a low negative-to-positive capacity ratio (N/P) of ~ 3.01 , and a lean electrolyte-to-capacity ratio (E/C) of 15 mL Ah^{-1} . The pouch cell was constructed by alternately stacking four NVO cathodes and five Zn anodes, yielding a designed capacity of $\sim 482 \text{ mAh}$. The ZT-AA-based pouch cell delivered stable charge-discharge performance over 22 cycles with good capacity retention (Figure S30), demonstrating that our bond-length engineering strategy is not only effective in coin cells but also promising for large-scale energy storage applications.

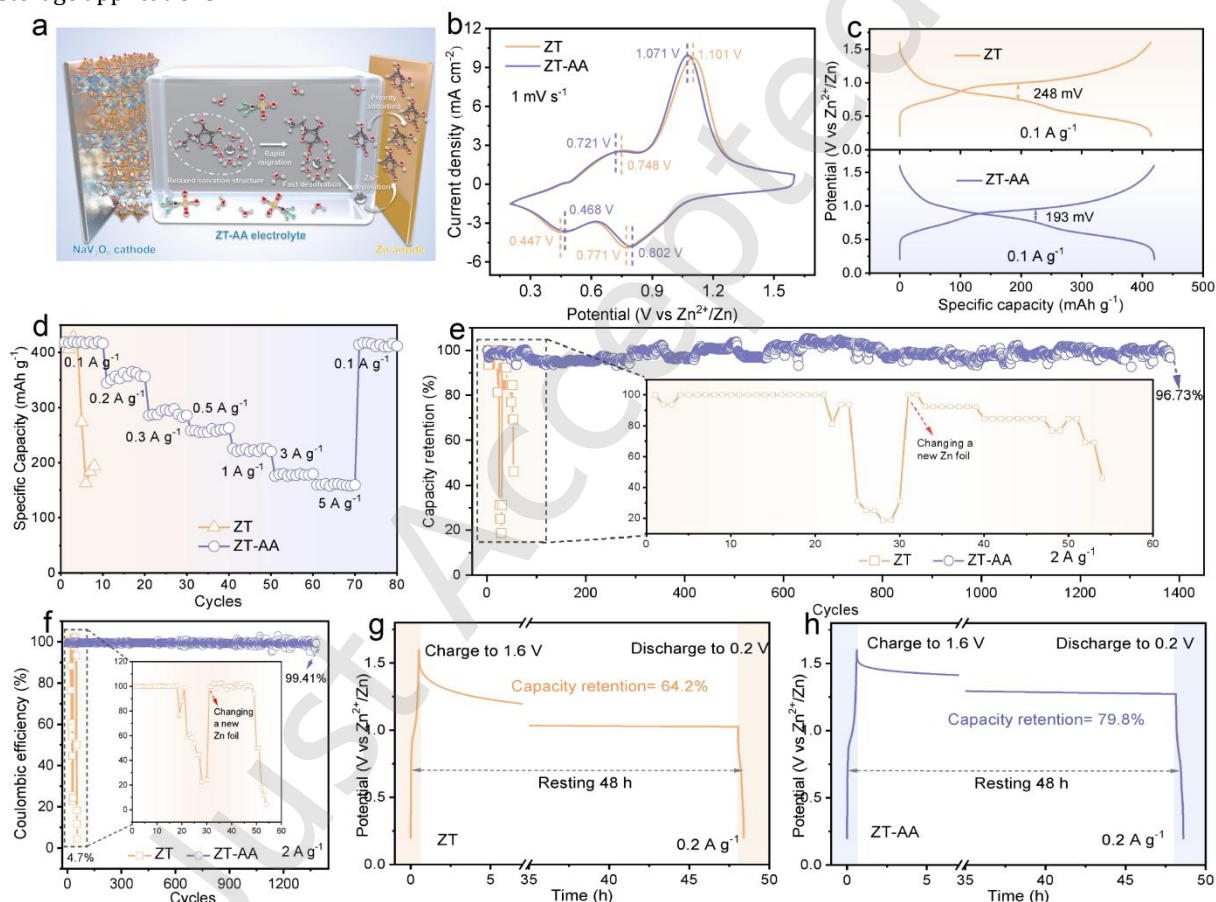


Figure 6. (a) Illustration of the Zn/NVO full-cell configuration. (b) CV responses, (c) GCD curves, (d) comparison of rate performance, (e) stability upon repeated cycling, (f) CE for Zn/NVO cells operated with ZT and ZT-AA. Comparison of self-discharge test results for Zn/NVO cells using (g) ZT versus (h) ZT-AA.

4 Conclusions

To conclude, we put forward an approach to stabilize Zn anode by tuning intrinsic reactivity of H_2O molecules, overcoming the performance trade-offs in aqueous electrolytes. Taking ZT-AA as an additive, experimental and theoretical results demonstrate that the combined electronic and steric hindrance effects from AA modulate the PSS, leading to an elongated $\text{Zn}^{2+}\text{-O}(\text{H}_2\text{O})$ bond and a stronger intramolecular O-H bond. This favorably facilitates Zn^{2+} desolvation kinetics and increases the energy barrier for H_2O reduction, thus inhibiting dendrite formation and HER. Moreover, AA molecules with hydroxyl and carbonyl groups can interfere with the hydrogen-bond framework of free H_2O and preferentially stick to the Zn electrode surface to build up a protective film, further suppressing HER. Driven by the tailored electrolyte properties, the symmetric Zn/Zn cell delivers an extended cycling lifespan under ultrahigh current density and areal capacity (100 mA cm^{-2} and 100 mAh cm^{-2}) with a remarkable DOD of 86.1%, substantially surpassing the performance of the ZT-based cell and most previously reported systems. Correspondingly, the assembled Zn/NVO full cell also possesses excellent cyclability, retaining 96.73% of its

capacity after ~1380 cycles with an average CE exceeding 99.41%. This bond-length engineering offers a promising direction for designing practical AZIBs.

Electronic Supplementary Material: Supplementary material (Raman spectra, FTIR spectra, EIS spectra, Tafel plots, and more electrochemical tests) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909134>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

L. Zhou: Methodology, investigation, data curation, writing-original draft. J. He and X. Shi: Methodology, data curation. S. Xie: Methodology, reviewing. X. Tong and M. Zhang: Methodology, reviewing. F. Cheng: Conceptualization, funding acquisition, writing-reviewing. X. Lu: Conceptualization, methodology, writing-reviewing and editing, supervision.

Use of AI statement

None.

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Electronic Supplementary Material

Bond-length engineering unlocks ultrahigh-DOD aqueous zinc batteries with relaxed solvation shell

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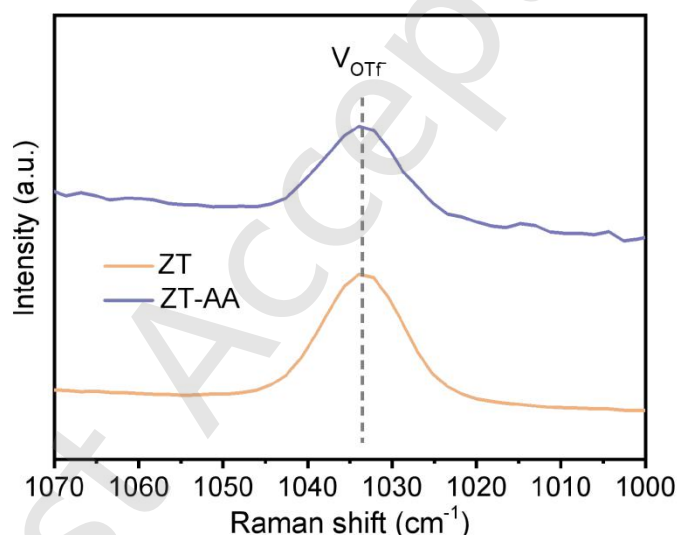


Figure S1. Raman spectra of ZT and ZT-AA.

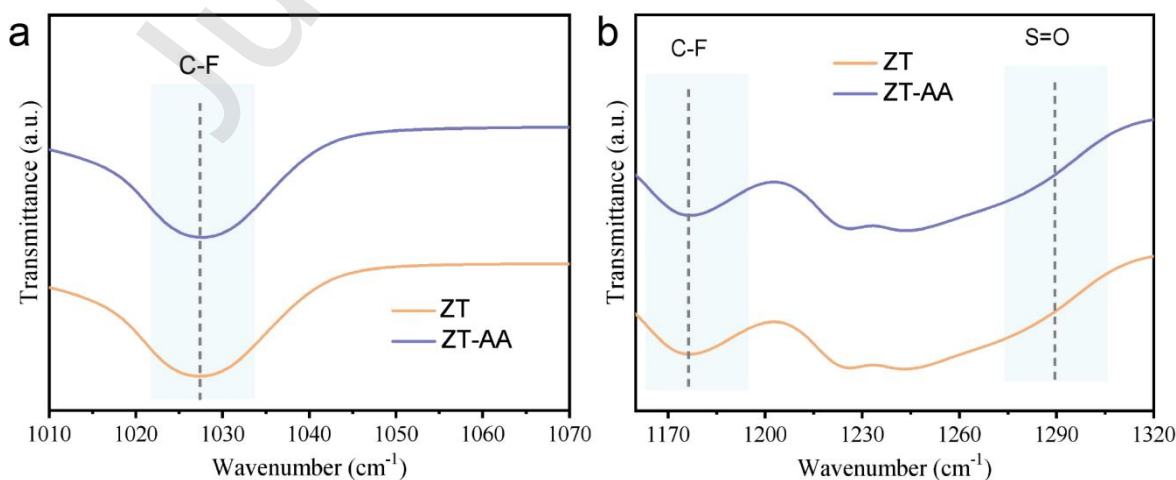


Figure S2. FTIR spectra of ZT and ZT-AA.

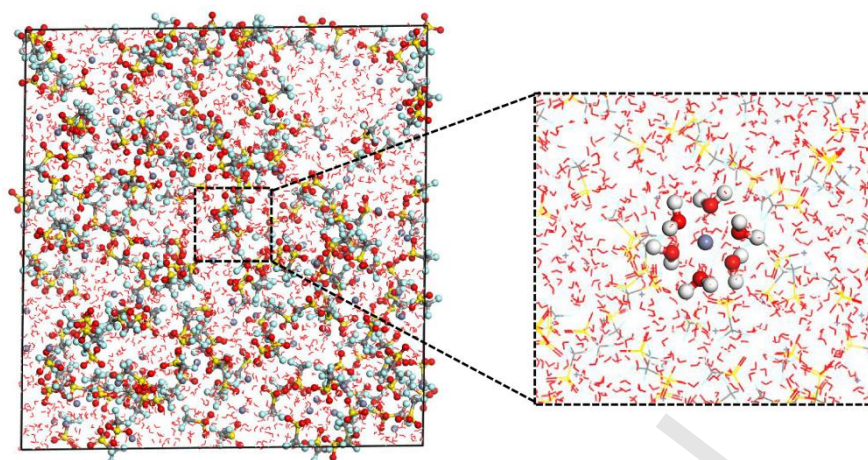


Figure S3. Snapshot of the MD simulation cell for ZT.

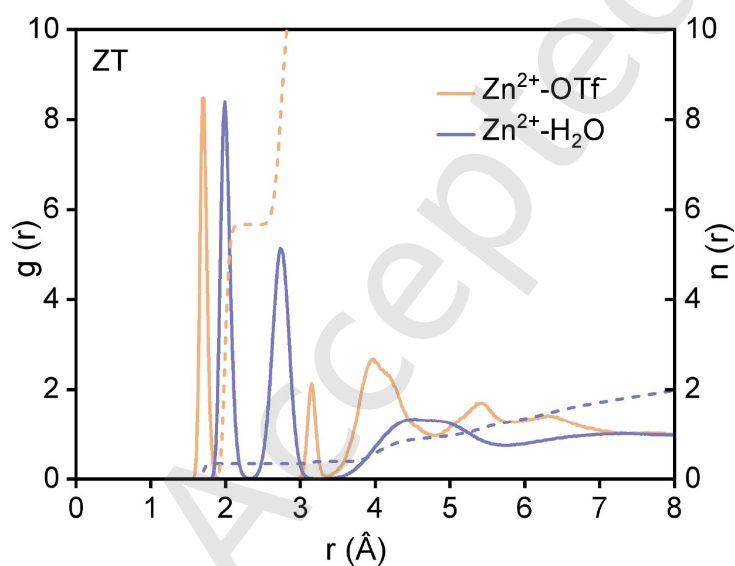


Figure S4. RDFs of Zn^{2+} -O (H_2O) and Zn^{2+} -O (OTf^-) pairs.

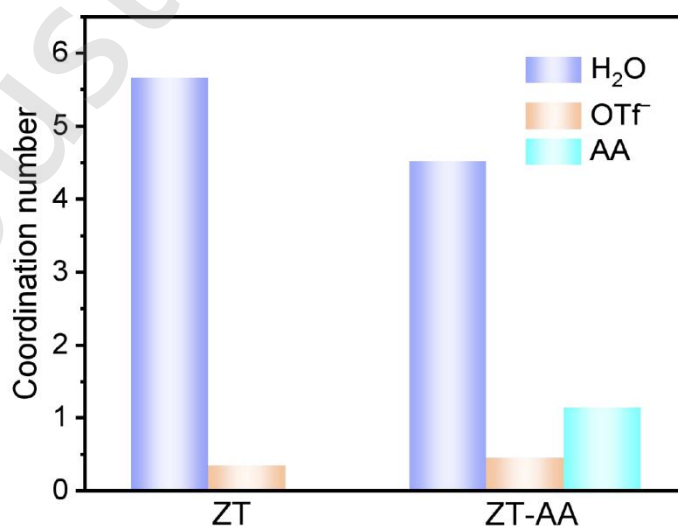


Figure S5. Coordination number of Zn^{2+} with different species in ZT and ZT-AA.

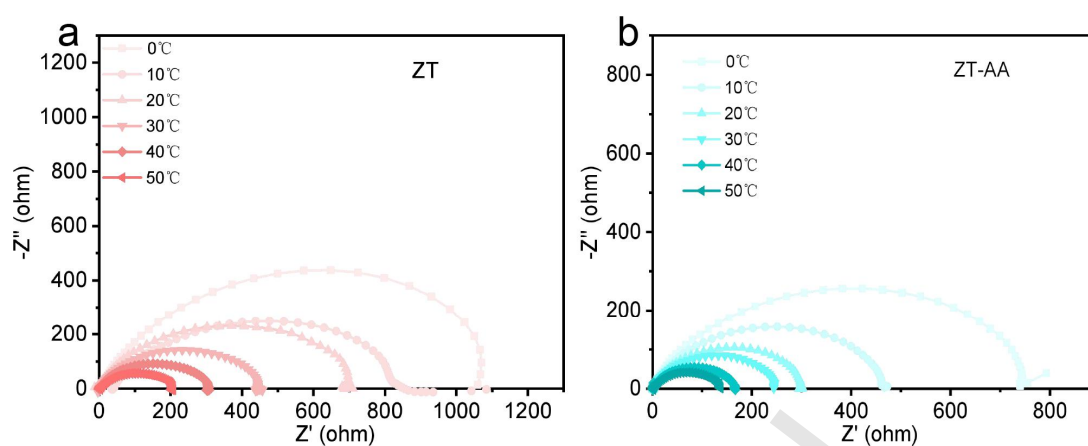


Figure S6. EIS spectra of symmetric Zn/Zn cells in (a) ZT and (b) ZT-AA tested under different temperatures.

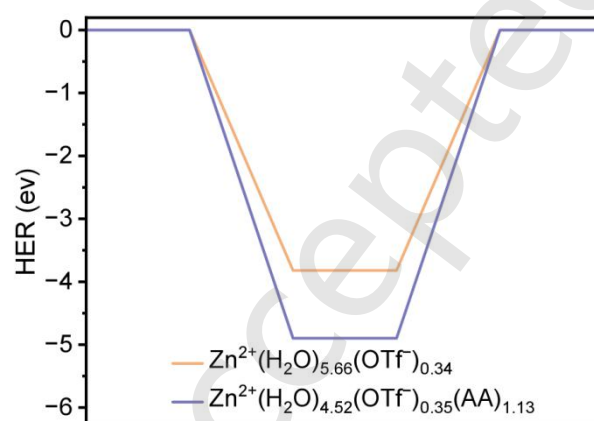


Figure S7. The energy barriers of HER for the different solvation structures of Zn^{2+} .

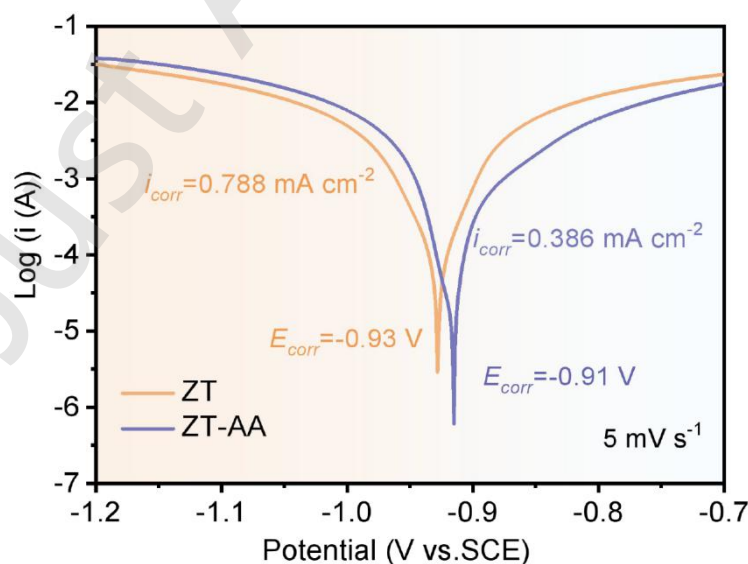


Figure S8. Tafel plots of Zn foil tested in ZT and ZT-AA at a scan rate of 10 mV s^{-1} .

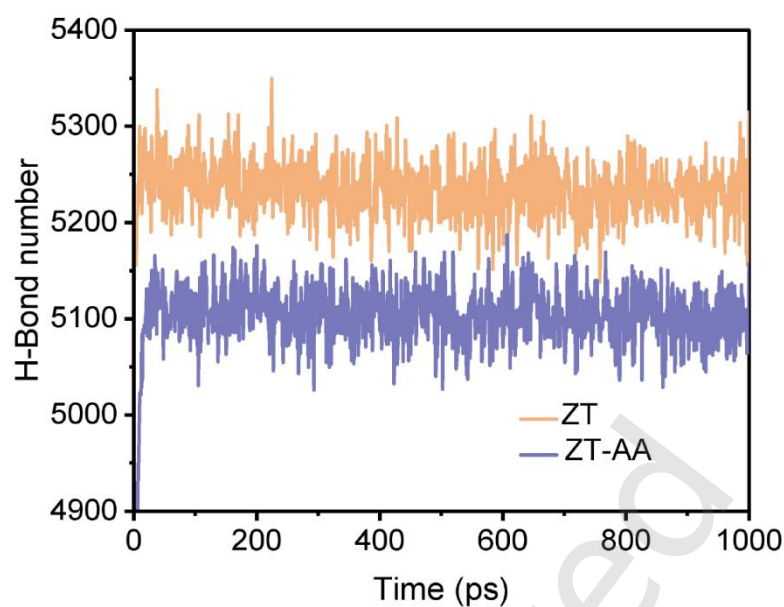


Figure S9. Number of hydrogen bond in different electrolytes simulated from MD.

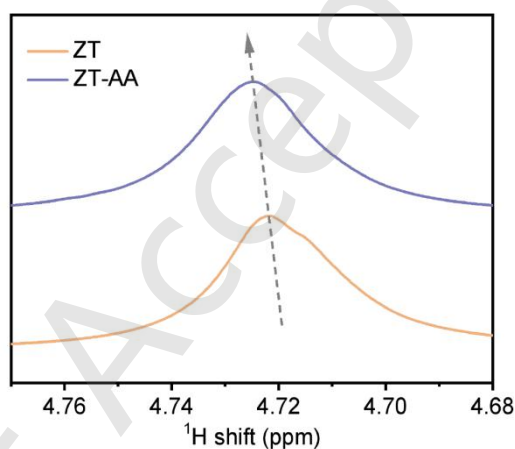


Figure S10. ^1H NMR spectra of H_2O from ZT and ZT-AA.

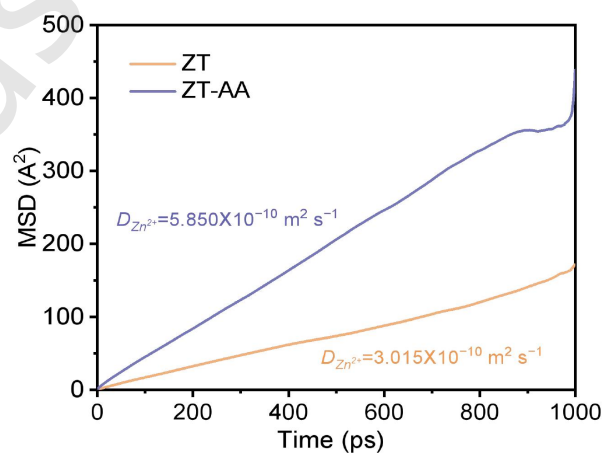


Figure S11. Diffusion coefficient of Zn^{2+} in ZT and ZT-AA simulated from MD.

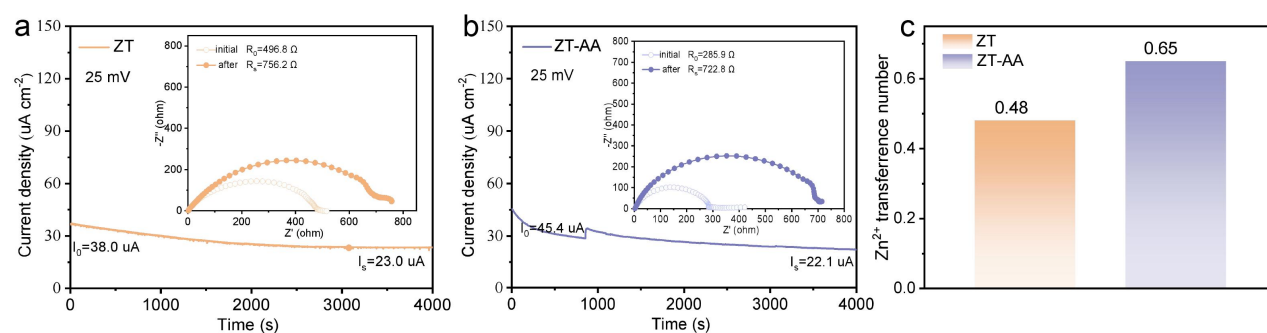


Figure S12. I-t curves of Zn cells in (a) ZT and (b) ZT-AA, insert shows Nyquist plots of Zn cells before and following polarization. (c) The Zn²⁺ transference number of Zn cells in ZT and ZT-AA.

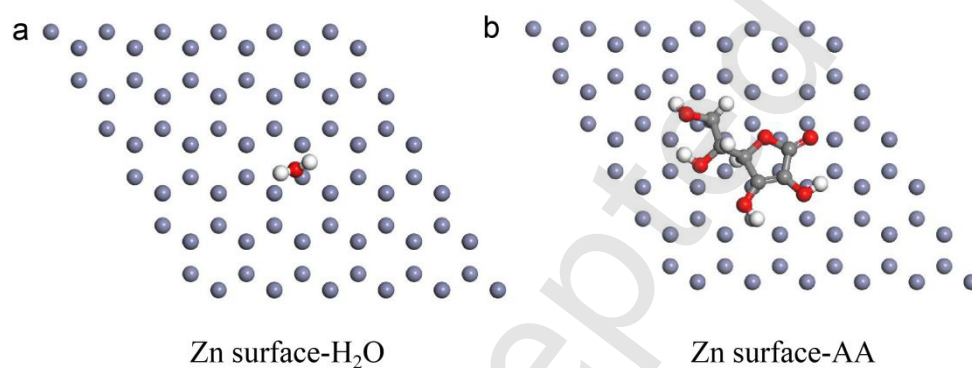


Figure S13. The calculated adsorption energy of (a) H₂O molecule and (b) AA molecule on Zn surface.

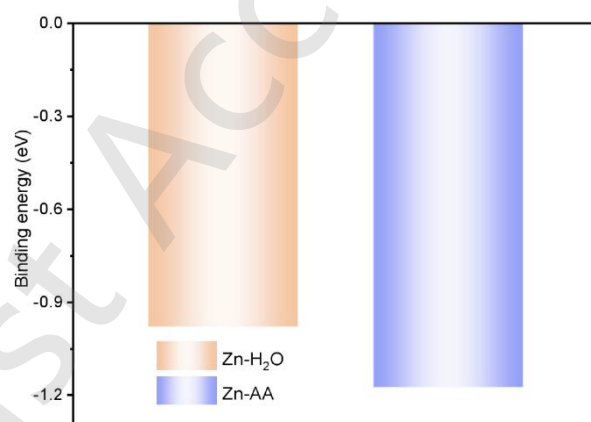


Figure S14. The compared adsorption energy of H₂O and AA molecules on Zn surface.

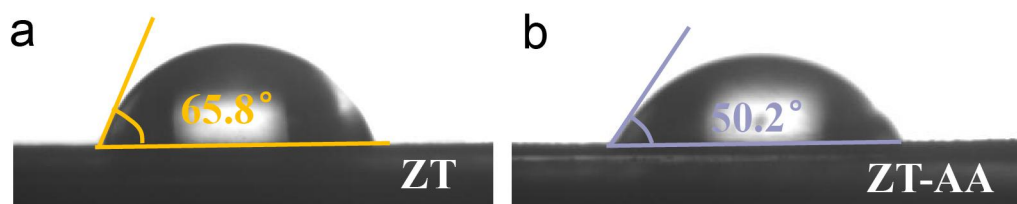


Figure S15. Contact angle of Zn foils with (a) ZT and (b) ZT-AA electrolytes.

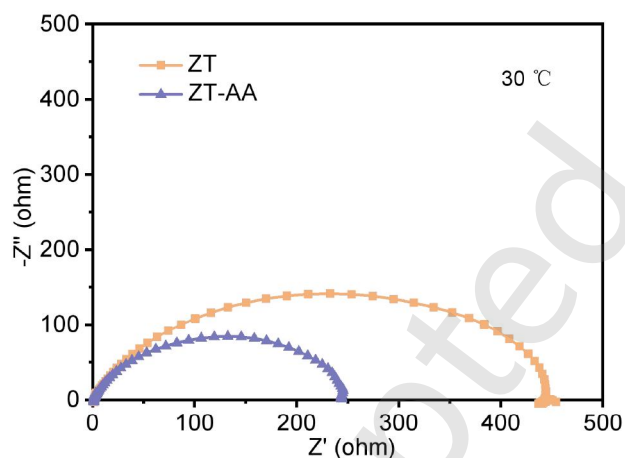


Figure S16. Nyquist plots of Zn/Zn cells in ZT and ZT-AA electrolytes.

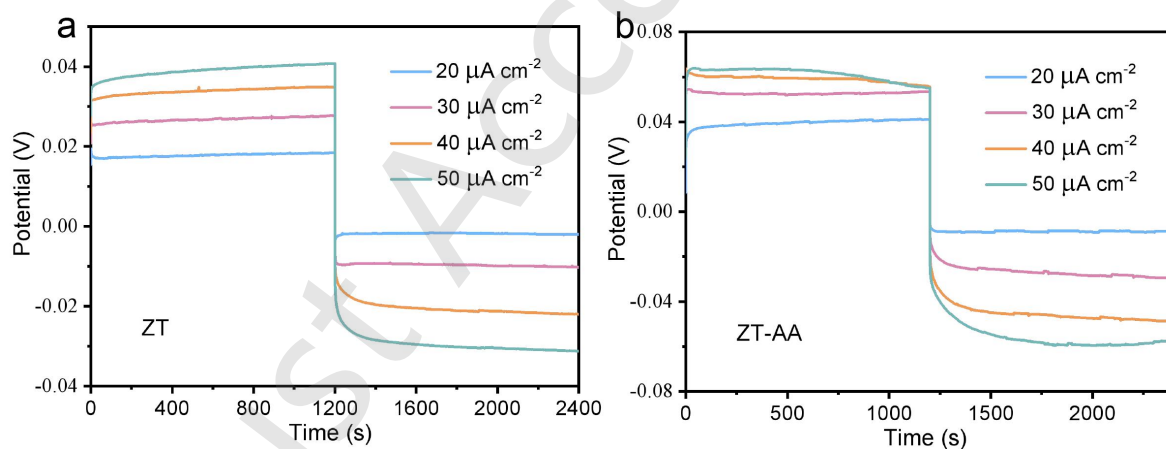


Figure S17. GCD curves of Zn/Zn symmetric cells in (a) ZT and (b) ZT-AA tested under different current densities.

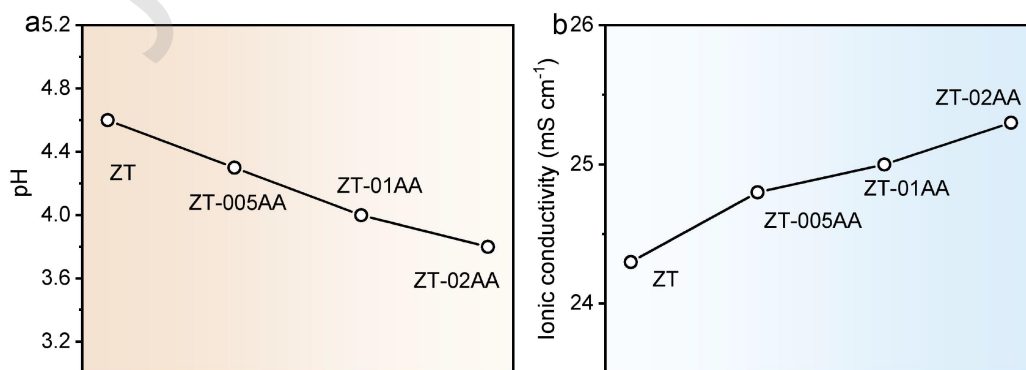


Figure S18. (a) pH and (b) ionic conductivity of electrolytes with 0.05M, 0.1M, 0.2M AA.

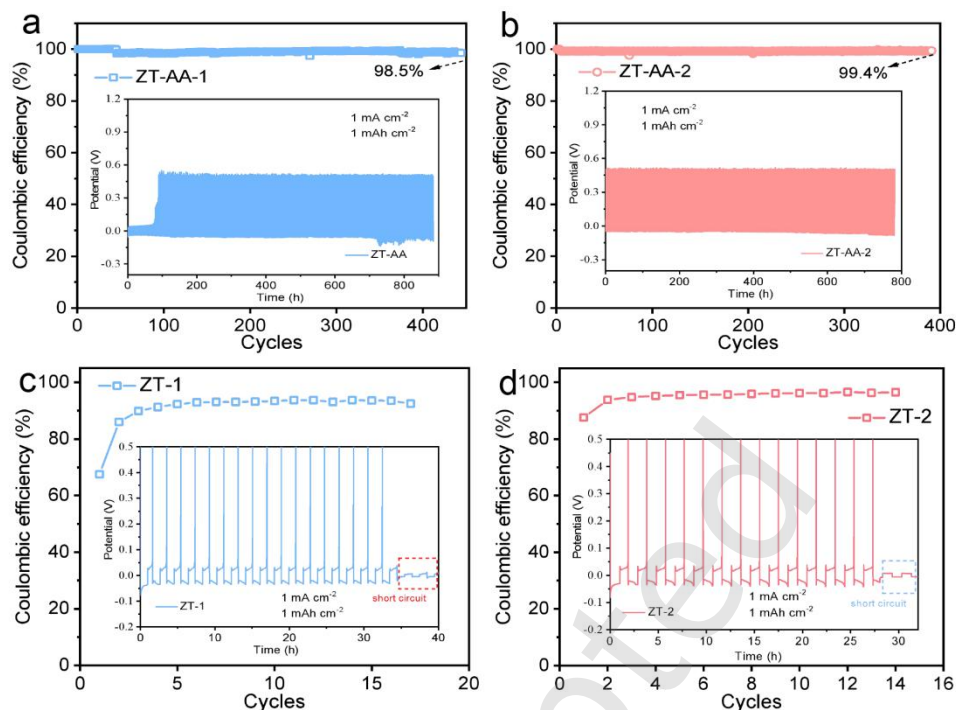


Figure S19. CE recorded for parallel Zn/Cu cells in (a,b) ZT-AA and (c,d) ZT electrolytes.

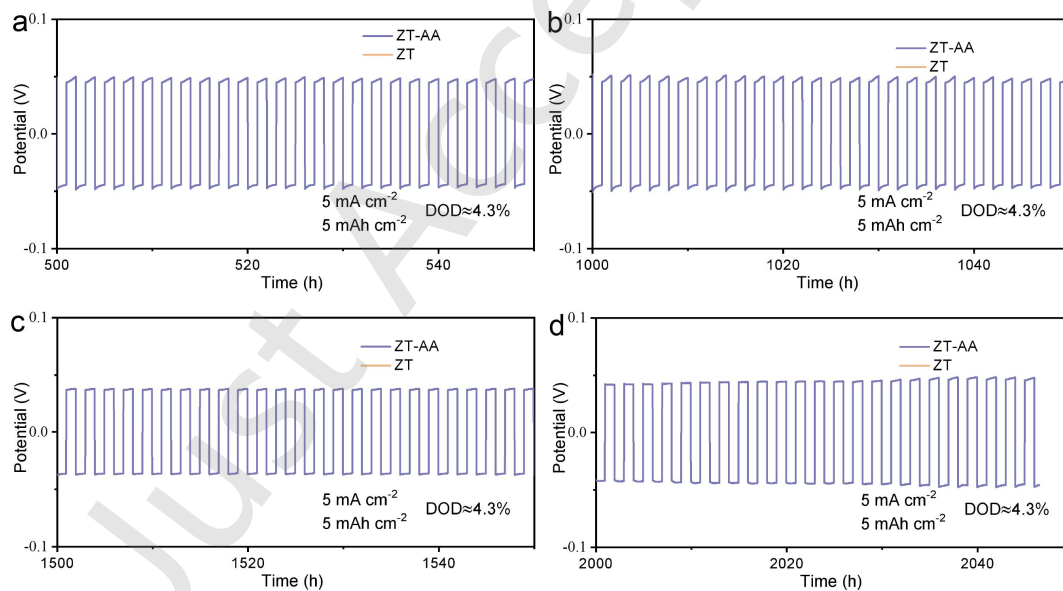


Figure S20. The locally enlarged curves of cycle performance of Zn/Zn cells in ZT and ZT-AA at 5 mA/mAh cm⁻² in Figure 5d.

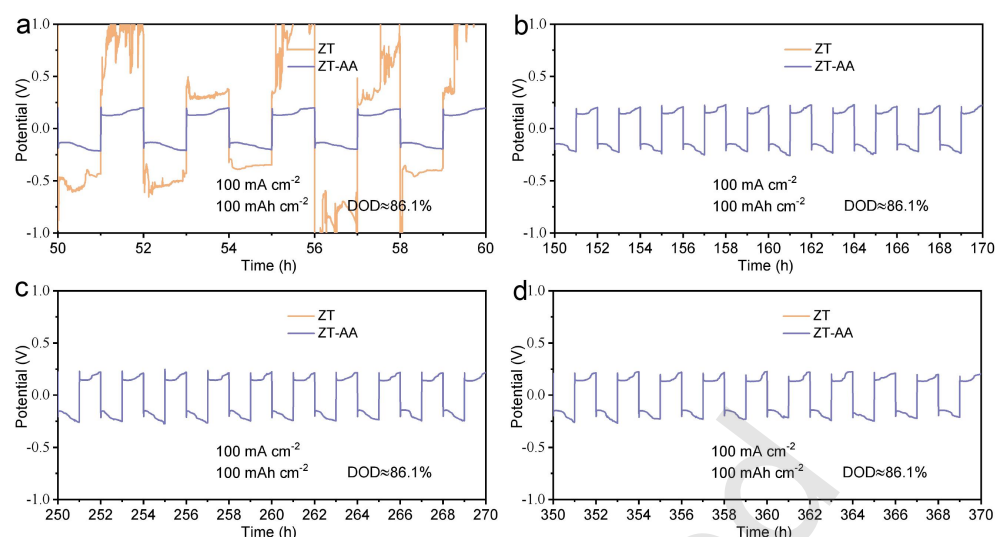


Figure S21. The locally enlarged curves of cycle performance of Zn/Zn cells in ZT and ZT-AA at 100 mA/mAh cm⁻² in Figure 5e.

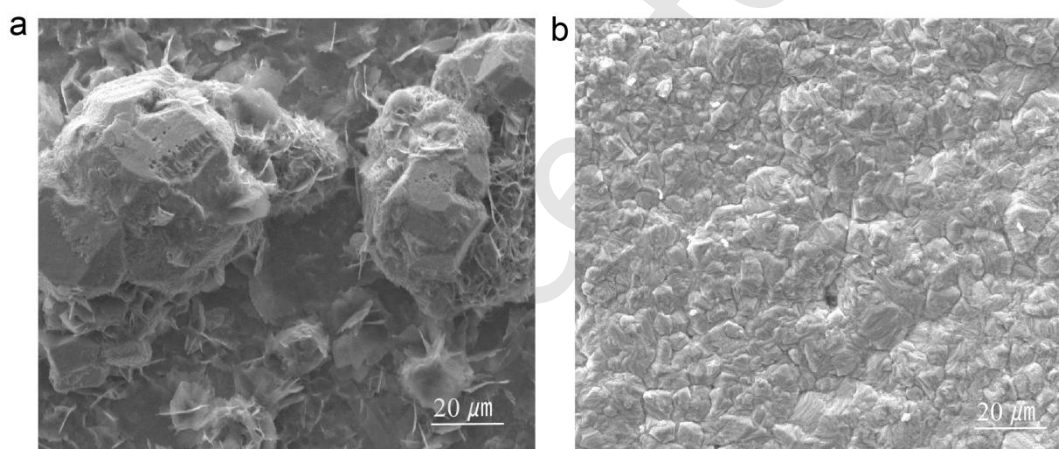


Figure S22. SEM images of Zn surfaces after cycling at (a) ZT and (b) ZT-AA electrolytes.

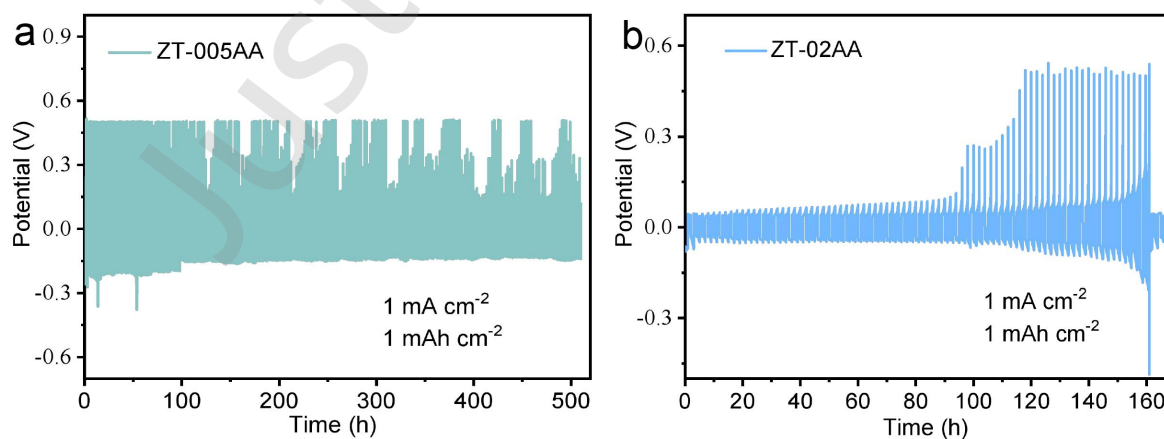


Figure S23. The cycling performance of Cu/Zn cells in (a) ZT-005AA and (b) ZT-02AA electrolytes.

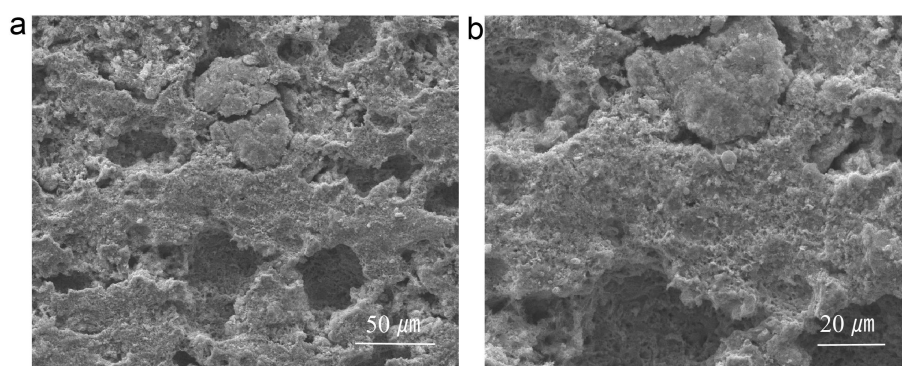


Figure S24. SEM images of Zn surface after cycling in ZT-02AA electrolyte.

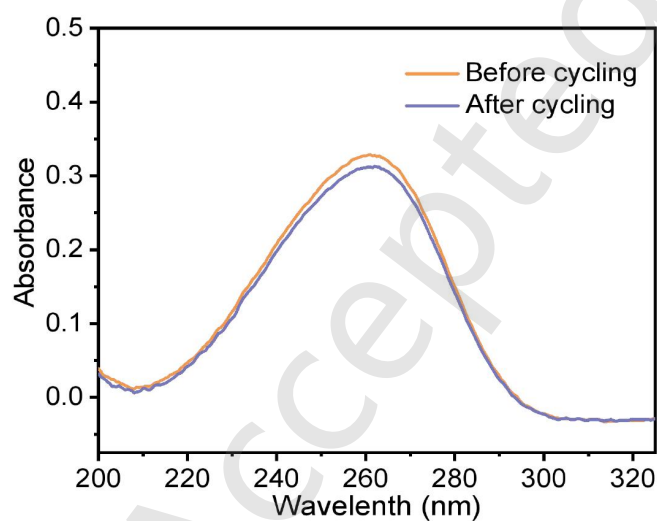


Figure S25. UV-vis absorption spectra of ZT-AA electrolyte before and after cycling.

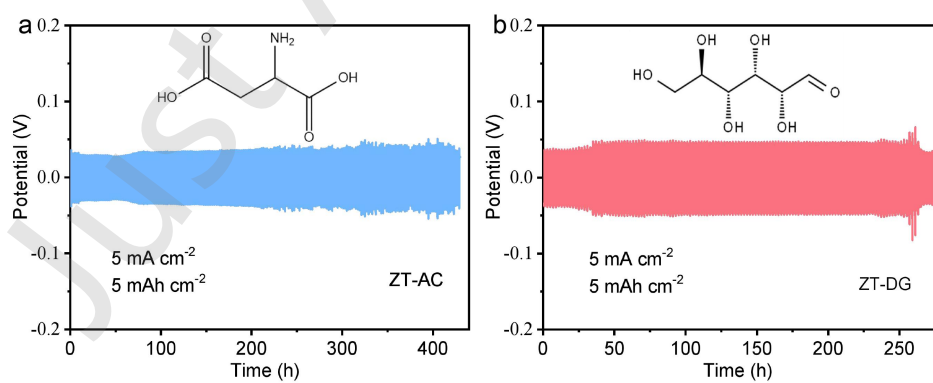


Figure S26. Cycling performance of Zn/Zn symmetric cells at 5 mA/mAh cm^{-2} in (a) ZT-AC and (b) ZT-DG electrolytes.

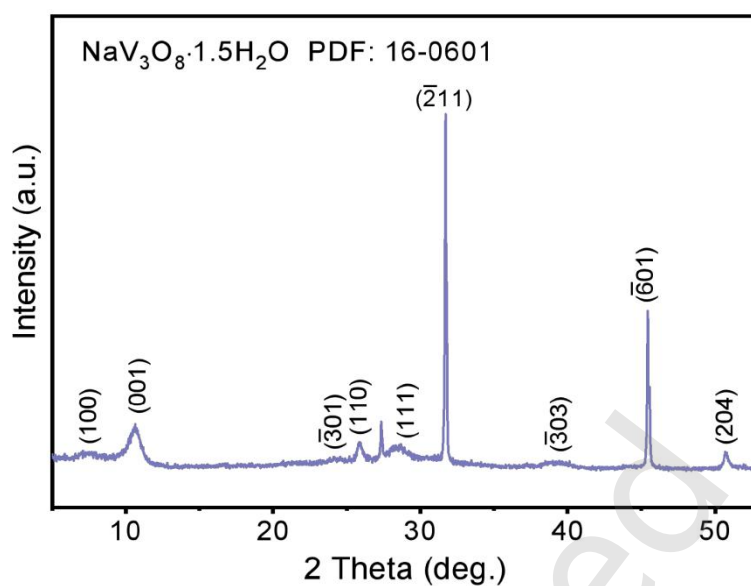


Figure S27. XRD pattern of NVO.

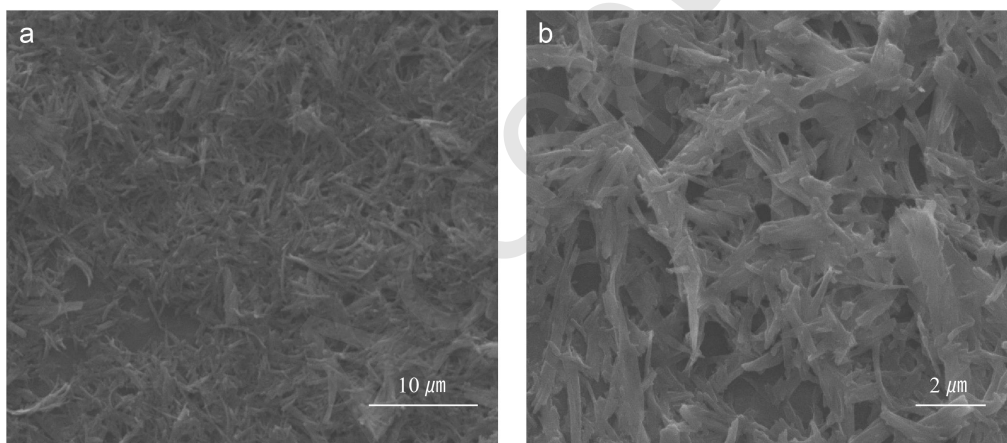


Figure S28. SEM images of NVO.

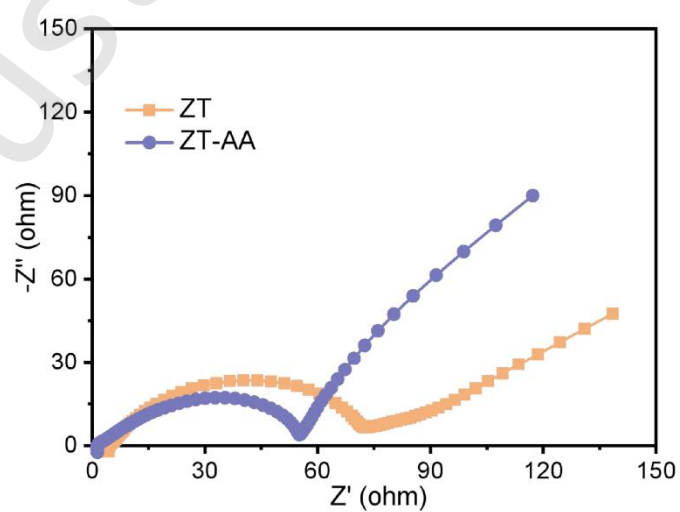


Figure S29. Nyquist plots of Zn/NVO cells in ZT and ZT-AA electrolytes.

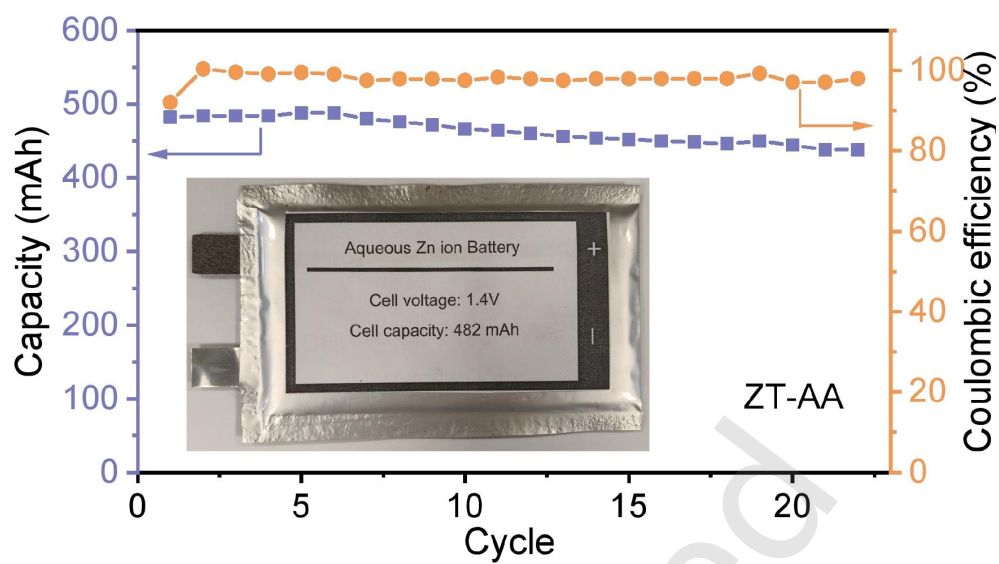


Figure S30. Cycling stability of Zn/NVO pouch cell, inset is the photograph of Zn/NVO pouch cell.