

ENERGY MATERIALS

Editor-in-Chief: Prof. Yuping Wu



 Open Access

ISSN 2770-5900 (Online)

Bare Zn





Ion sieving function of MoS₂ and Alg-Zn hybrid coating endows high stability of Zn anode for aqueous Zn-ion batteries

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Keywords:

Aqueous Zn-ion batteries, uniform Zn deposition, zinc alginate, molybdenum disulfide, cycle stability

Citation:

He, Y.; Zhang, Z.; Jin, K.; Yuan, X.; Sun, Z.; Fan, W.; Yuan, W.; Han, P.; Fu, L.; Wu, Y. Ion sieving function of MoS₂ and Alg-Zn hybrid coating endows high stability of Zn anode for aqueous Zn-ion batteries. *Energy Mater.* 2026, 6, 600002. <https://dx.doi.org/10.20517/energymater.2025.126>

Received: 7 Aug 2025

First Decision: 29 Aug 2025

Revised: 7 Oct 2025

Accepted: 23 Oct 2025

Published: 16 Jan 2026

Academic Editor:

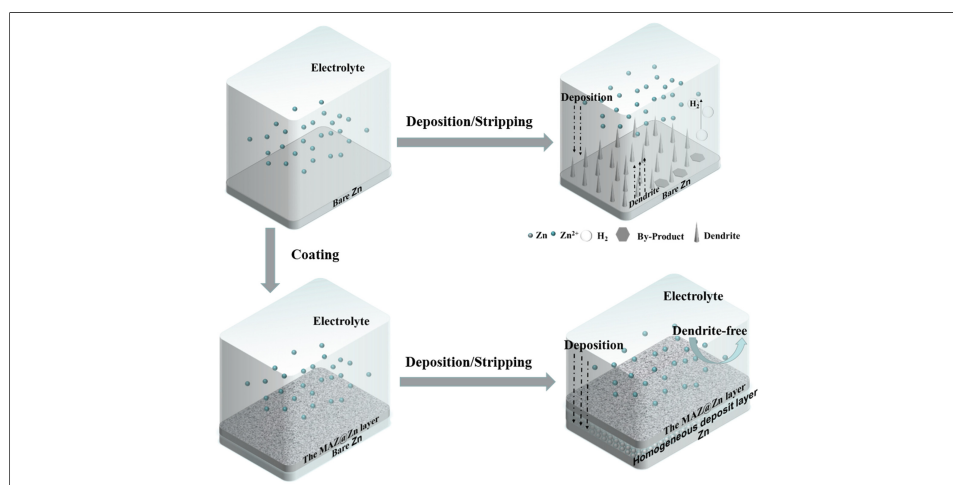
Jiaqi Huang

Copy Editor:

Fangling Lan

Production Editor:

Fangling Lan



Abstract

Aqueous Zn-ion batteries (AZIBs) have emerged as promising energy storage systems due to their high safety, low cost, and environmental friendliness. However, the practical application of zinc metal anodes is hindered by challenges such as Zn dendrite growth and side reactions, which degrade the cycle performance and energy efficiency of AZIBs. To address these issues, a facile and functional coating composed of zinc alginate gel (Alg-Zn) and 2H-molybdenum disulfide (2H-MoS₂) was used to modify the Zn anode (MAZ@Zn). Combined experimental and theoretical investigations reveal that, in addition to the Zn²⁺ guiding effect of ion conductive Alg-Zn, the 2H-MoS₂ functions as an ion sieve. This facilitates the fast Zn²⁺ migration and even distribution because of the lower ion migration energy along the MoS₂ surface, ensuring fast Zn²⁺ diffusion in the MAZ@Zn coating and uniform Zn deposition. Moreover, the barrier effect of MoS₂ against H₂O helps suppress side reactions such as hydrogen evolution, thereby further enhancing the interfacial

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stability of the Zn anode. As a result, the MAZ@Zn symmetric cells exhibit excellent cyclic stability, achieving a lifespan of 880 h at 1 mA cm⁻² and 1 mAh cm⁻², with low voltage polarization and low charge transfer energy. In contrast, the bare Zn anode only sustains 150 h of cycling under identical conditions. In Zn//sodium vanadate full batteries, the MAZ@Zn anode demonstrates outstanding performance, retaining 88.4% of its capacity after 1,000 cycles at 4 A g⁻¹. This work offers a simple and effective strategy for developing high-performance Zn anodes for long-life AZIBs.

INTRODUCTION

Aqueous Zn-ion batteries (AZIBs) are promising candidates for next-generation energy storage devices, owing to the inherent advantages of zinc-metal anodes^[1,2]. These advantages include low cost^[3], abundant resource availability, compatibility with aqueous electrolytes, high theoretical capacity (820 mAh g⁻¹)^[4], and an appropriate redox potential (-0.76 V vs. the Standard Hydrogen Electrode (SHE))^[5]. This specific potential window effectively inhibits hydrogen evolution reactions (HERs) at the Zn/electrolyte interface, thereby ensuring electrochemical stability^[6]. Despite these benefits, the practical use of zinc metal anodes remains challenging, such as the formation of Zn dendrites during the electroplating process^[7], side reaction with water, which can deteriorate the battery performance^[8,9].

In response to these challenges, great efforts have been made to enhance the performance of AZIBs^[10-12]. Strategies include designing three-dimensional Zn anodes and Zn alloys^[13,14], employing gel or solid-state electrolytes^[15-18], and introducing electrolyte additives^[19-21]. Among these, the development of artificial coatings to inhibit the growth of Zn dendrites has emerged as an effective and practical solution^[22-25].

The coating surface reported in the literature can be simply classified into inorganic and organic coatings. Inorganic coatings mainly consist of metals^[26,27] (In, Ag, *etc.*), metal oxides^[28-31] (TiO₂, ZnO, Al₂O₃, *etc.*) and carbon materials^[32,33]. They can inhibit dendrite growth but tend to increase the resistance at the electrolyte-electrode interface^[34], adversely affecting overall battery performance. Organic materials with polar groups^[35-38] (meta-organic frameworks, polyamide, poly (2-vinylpyridine)) - although capable of guiding Zn²⁺ migration - often suffer from high polarization. This significantly affects the rate performance of the batteries^[39,40]. Therefore, it is of great significance to explore the surface coating of the Zn anode in order to reduce the disadvantages of inorganic coating or organic coating with facile preparation.

Zinc alginate (Alg-Zn) demonstrates potential as an effective organic coating material, exhibiting high ionic conductivity (1.83 × 10⁻² S cm⁻¹)^[41]. By binding with free water molecules, it significantly improves electrolyte wettability. Furthermore, the Zn-affinity carboxyl and ether groups within its polyanionic framework enable guided two-dimensional Zn²⁺ diffusion^[41,42]. In our prior study, an engineered mixed ionic-electronic conductive coating (Alg-Zn + AB@Zn) - comprising Alg-Zn gel and acid-treated carbon black (AB) - was developed to enhance Zn anode stability^[43]. The prepared mixed coating not only promoted uniform Zn nucleation, but also effectively reduced the nucleation overpotential of Zn. The Alg-Zn + AB@Zn symmetrical cell exhibited a low polarization voltage (47 mV) and achieved long-term cycle stability for 500 h. The assembled Zn//activated carbon (AC) full battery was able to cycle over 10,000 times at 10 A g⁻¹ and 16,000 times at 20 A g⁻¹, with a capacity retention rate close to 100%. This performance surpassed other reported Zn//AC full batteries at high current densities. However, due to the electrical conductivity of AB, Zn²⁺ deposition occurred on the coating layer, and the cycle performance of the artificially mixed conductor coating-modified Zn still required further improvement.

MoS₂ demonstrates a strong capability for promoting the homogeneous deposition of Zn²⁺ and inhibiting dendrite formation. Wang *et al.* prepared a large-area quasi-single-crystal 2D material (molybdenum disulfide, MoS₂) as a substrate using an epitaxial phase-conversion method^[44]. The results showed that the edge of 2H-MoS₂ exhibited a stronger Zn deposition induction effect than the base plane, which inhibited the growth of Zn dendrites. Bhoyate *et al.* developed a 2D MoS₂ coating on a Zn anode via electrochemical deposition^[45]. The coated MoS₂ layer formed a vertically oriented structure that facilitated the easy flow of Zn²⁺ and ensured a uniform electric field distribution on the anode. This led to uniform stripping and plating of Zn²⁺, preventing dendrite growth and complex side reactions. When tested at a current density of 2.5 mA cm⁻² and a capacity of 0.416 mAh cm⁻², the MoS₂-Zn symmetric cell cycled for 175 h with a polarization voltage of 120 mV. The MoS₂-Zn//MnO₂ full battery achieved a reversible specific capacity of 638 mAh g⁻¹ at 0.1 A g⁻¹ and maintained stable cycle performance over 2,000 cycles. However, the preparation of the MoS₂ coating was complex.

Here, we use a simple drop-coating method to obtain a hybrid gel-conductive coating consisting of 2H-MoS₂ and Alg-Zn (MAZ@Zn). When sodium alginate is immersed in the 2 M ZnSO₄ electrolyte, it interacts with Zn²⁺ to form Alg-Zn, which can transfer Zn²⁺. This creates a “channel” where Zn²⁺ migration is guided and confined by the carboxylate groups of alginate^[43-46]. The superimposed or cross-linked flexible channels formed by MoS₂ facilitate rapid Zn²⁺ transfer and guide the homogeneous deposition of Zn²⁺ on the surface of zinc foil beneath the coating^[47]. First-principles calculation reveals that Zn²⁺ exhibits reduced migration energy on the 2H-MoS₂ (002) interface, making this surface an adept sieve for these ions. Such a design guarantees the homogeneous distribution of Zn²⁺ on the electrode surface, curbing the emergence of Zn dendrites effectively. In addition, the barrier effect of MoS₂ against H₂O may help suppress side reactions such as hydrogen evolution, further enhancing interfacial stability. The symmetric cell constructed with MAZ@Zn achieves a cycle life of 880 h at a current density of 1 mA cm⁻², while the unmodified bare Zn anode only shows a cycle life of 150 h. Compared with previous studies [Supplementary Table 1], the MAZ@Zn anode exhibits competitive cyclic stability (880 h), which can be attributed to the uniform distribution of Zn²⁺ on the electrode surface, effectively inhibiting the formation of Zn dendrites. In addition, the full MAZ@Zn//Zn// sodium vanadate (NVO) battery retained 88.4% of its capacity after 1,000 cycles at a current density of 4 A g⁻¹.

EXPERIMENTAL

Preparation of Zn anode modified with a composite layer of Alg-Zn and MoS₂

First, sodium alginate and commercial MoS₂ powder were initially blended in deionized water at room temperature (25 °C). The mass ratios for the sodium alginate and MoS₂ powder were varied as 3:7, 1:1, and 7:3, while the sodium alginate-to-deionized water mass ratio was maintained at 1:50^[43]. This blend was then subjected to magnetic stirring for a duration of 12 h to ensure thorough mixing. Leveraging sodium alginate's properties as a binder, the slurry was carefully mixed until it achieved a viscous yet flowable consistency. Following this, the slurry was meticulously drop-cast and uniformly spin-coated onto a polished and smooth Zn plate to ensure even distribution. The coated Zn plate was subsequently immersed in 2 M ZnSO₄ solution, which facilitated the ion-exchange process between sodium alginate and zinc alginate in the coating. Specifically, Zn²⁺ ions replaced Na⁺ ions in the carboxylate groups of the alginate chains, forming coordination bonds that give rise to the characteristic “egg-box” structure of Zn alginate. This crosslinking with divalent Zn²⁺ significantly enhances the stability of the alginate matrix compared with monovalent Na⁺, thereby reducing solubility and improving resistance against swelling or dissolution in aqueous electrolytes. Upon removal from the solution, a composite coating comprising Zn alginate and MoS₂ was formed on the Zn surface. This resulted in the formation of three distinct coated materials: M7AZ3@Zn, MAZ@Zn, and M3AZ7@Zn, each indicating the respective ratios of MoS₂ to Alg-Zn in the coating.

Preparation of sodium vanadate (NVO) cathode material

Initially, 1.5 g of commercial V_2O_5 powder was mixed into 50 mL of 2 M NaCl solution. This mixture was then subjected to ultrasonication to achieve a uniform dispersion. Following this, the solution was continuously stirred for 72 h. When the solution acquired an orange hue, it was filtered, and the resulting residue was meticulously rinsed with deionized water and anhydrous ethanol. The next step involved vacuum drying the residue at 80 °C for 12 h. For the composition of the active material, NVO, conductive carbon black, and binder PTFE were proportionally weighed in a mass ratio of 7:2:1. These components were then ground together and dissolved in anhydrous ethanol to form a slurry, which was subsequently exposed to a baking lamp. The obtained material was then pressed onto a stainless-steel mesh current collector using a roller press, with the areal loading of the active material controlled at approximately 4.5 mg cm^{-2} .

Electrochemical measurements

In the electrochemical tests for Zn/Zn symmetric cells, two polished Zn foils, each 100 μm thick and 15 mm in diameter, were used as electrodes. To assemble the battery, 70 μL of a 2 M ZnSO_4 electrolyte solution was added into a CR2025 button cell using a pipette. A glass fiber separator (GF/A, Whatman) was used to separate the electrodes in the battery. For the Zn/Cu asymmetric cells, the process was the same as for the Zn/Zn cells. However, one Zn foil was replaced with a Cu foil, which was 32 μm thick and 15 mm in diameter. For the Zn//NVO full batteries, a Zn foil was used as the anode, and NVO material was used as the cathode. A glass fiber membrane was used as the separator, similar to the other battery types. The electrolyte was a mixture of 70 μL of 2 M ZnSO_4 and 1 M Na_2SO_4 . All parts were carefully assembled into a CR2025 button battery format for testing. The ionic conductivity (σ) of the electrolytes was derived from electrochemical impedance spectroscopy (EIS) data collected over 10 mHz–100 kHz on Zn//Zn symmetric cells. A custom-designed fixture maintained a fixed electrode gap to ensure reproducible conductivity estimates. The conductivity was calculated using:^[48]

$$\sigma = \frac{L}{R_b S} \quad (1)$$

where R_b is the bulk resistance taken from the high-frequency intercept on the real axis of the Nyquist plot, L denotes the coating thickness, and S is the effective electrode-electrolyte contact area. The contribution of the MAZ coating (R_{MAZ}) was isolated by subtraction: $R_{\text{MAZ}} = R_{\text{total}} - R_{\text{glass-fiber}}$.

The Zn^{2+} transference number ($t_+(\text{Zn}^{2+})$) was obtained using the Evans method:^[49]

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

where ΔV is the applied polarization voltage, I_o and I_s are the initial and steady-state current densities, respectively, and R_o and R_s are the charge-transfer resistances measured before and after polarization. Chronoamperometry was conducted under a constant 25 mV bias, which corresponds to ΔV .

The desolvation activation energy (E_a) was determined by fitting the charge-transfer resistance (R_{ct}) measured at different temperatures to an Arrhenius-type relation:^[50]

$$\frac{1}{R_{ct}} = A \exp\left(-\frac{E_a}{RT}\right) \quad (3)$$

where R_{ct} is the interfacial charge-transfer resistance, A represents the pre-exponential factor, T denotes the absolute temperature, R indicates the gas constant, and E_a signifies the activation energy for desolvation.

Characterization

Fourier Transform Infrared Spectroscopy (FTIR) data were collected using a Nicolet IS50 spectrometer with an Attenuated Total Reflection attachment. The crystal structure was analyzed with an X-ray diffractometer (XRD, D/MAX-IIA, Rigaku) using Cu-K α radiation ($\lambda = 0.15406$ nm) and a scanning angle (2θ) range from 10° to 90° . Scanning Electron Microscopy (SEM) images were taken at 10 kV or 15 kV using a Phenom XL-70 instrument. An optical microscope was used to examine the samples at a microscopic level. All samples were analyzed using the YM520R optical microscope from Yueshi Company in Suzhou, China. The cycling performance of the batteries was tested using the LAND battery test system (CTA2001A, Wuhan Land Electronics Co., Ltd., China). Cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and EIS tests were conducted with the CHI660E electrochemical workstation (Shanghai, China). The EIS range was 0.01–100 kHz.

Computational details

In the density functional theory (DFT) calculation, we initially optimize the geometry structures of Zn (002) surface and 2H-MoS₂ (002) surface until the force on each atom is reduced to less than 1×10^{-4} eV Å⁻¹ and obtain the total energy of the Zn and MoS₂ surface. Then, Zn atoms are put on each selected adsorption site. Following the geometry optimization of Zn (002) [or MoS₂ (002)] surface with adsorption atoms, the total energy of the adsorption system is obtained. We then calculate the adsorption energy using:^[51]

$$E_{abs} = E(\text{surface} + \text{Zn}) - E(\text{surface}) - E(\text{Zn}) \quad (4)$$

where $E(\text{Zn})$ indicates the total energy of a Zn atom. $E(\text{surface})$ is the total energy of the clean surface, and $E(\text{surface} + \text{Zn})$ is the total energy of the surface with the adsorbed Zn atom.

All the DFT calculations are performed using the CP2K code package. In these calculations, the molecular orbitals of the valence electrons were expanded using a hybrid double-zeta valence polarisation (DZVP)-Molecular Optimization(MOLOPT)-Goedecker-Teter-Hutter (GTH) Gaussian basis set, with plane waves expanded to a cutoff of 500 Rydberg^[52]. The GTH pseudopotentials within the framework of the local density approximation functional are employed to describe the atomic core electrons, while the DFT-D3 method accounts for van der Waals interactions^[53,54].

RESULTS AND DISCUSSION

This study investigated the impact of Alg-Zn to MoS₂ mass ratios on the performance of Zn anode electrodes. Raman spectroscopy [Supplementary Figure 1A] confirmed that the experimental MoS₂ was in the 2H phase^[55]. Different compositions of Alg-Zn and MoS₂ coatings were prepared, with mass ratios of 3:7 (M3AZ7), 1:1 (MAZ), and 7:3 (M7AZ3). The surface morphology of these coatings, along with that of bare Zn, was examined using SEM. As shown in Figure 1, bare Zn exhibits a relatively flat surface interspersed with fine scratches, likely resulting from the polishing process of the Zn foil [Figure 1A]^[56]. However, M7AZ3@Zn, MAZ@Zn and M3AZ7@Zn consist of irregular aggregated particles with dense surfaces and uniform distribution of MoS₂ particles [Figure 1B–D], which are conducive to electrolyte penetration and

provide an improved interface for electrochemical reactions^[43].

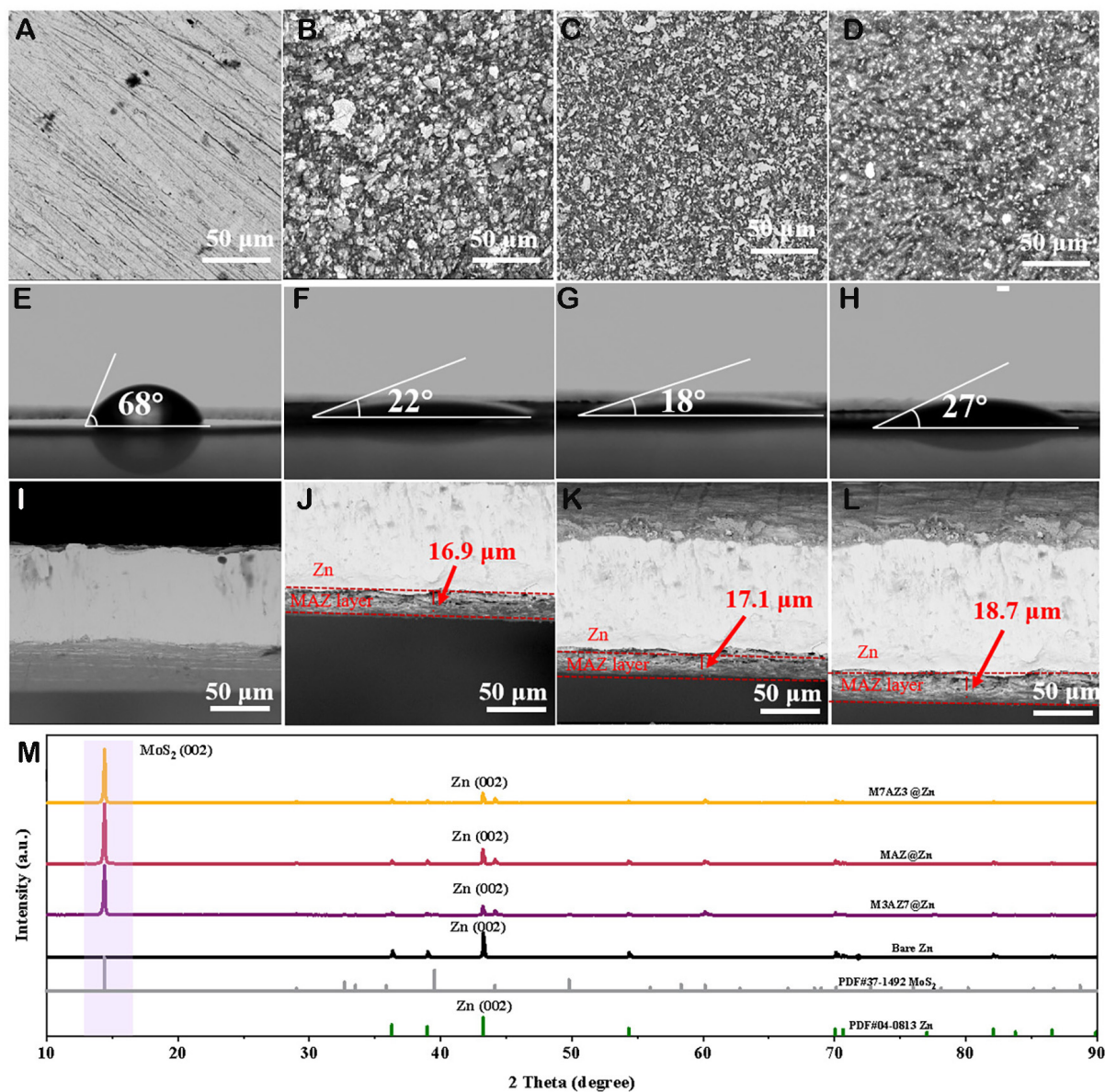


Figure 1. Physical property characterization of Zn anodes. SEM images of (A) bare Zn, (B) M7AZ3@Zn, (C) MAZ@Zn, and (D) M3AZ7@Zn. Contact angle measurements of (E) bare Zn, (F) M7AZ3@Zn, (G) MAZ@Zn, and (H) M3AZ7@Zn. Cross-sectional SEM images of (I) bare Zn, (J) M7AZ3@Zn, (K) MAZ@Zn, and (L) M3AZ7@Zn. (M) Initial XRD patterns of bare Zn, M7AZ3@Zn, MAZ@Zn, and M3AZ7@Zn.

The efficacy of the MAZ@Zn coating is further evaluated through contact angle measurements conducted in 2 M ZnSO₄ aqueous solution, as depicted in Figure 1E-H. Remarkably, while the bare Zn sample exhibits a contact angle of 68° [Figure 1E], the coated samples of M7AZ3@Zn, MAZ@Zn, and M3AZ7@Zn demonstrate significantly lower contact angles of 22°, 18°, and 27°, respectively [Figure 1F-H]. The contact angles of M7AZ3@Zn and M3AZ7@Zn are larger than that of MAZ@Zn, but for different reasons. For M7AZ3@Zn, the higher contact angle is due to its larger fraction of hydrophobic MoS₂. For M3AZ7@Zn, despite having more Alg-Zn, the contact surface with the electrolyte is insufficient, leading to a similarly higher contact angle. For MAZ@Zn, the homogeneous mixing of MoS₂ with Alg-Zn creates a hybrid structure that facilitates electrolyte penetration, thereby resulting in a smaller contact angle. Further cross-sectional SEM observations reveal that the coating thicknesses of M7AZ3@Zn, MAZ@Zn, and M3AZ7@Zn are 16.9 μm, 17.1 μm, and 19.7 μm, respectively, as shown in Figure 1J-L.

To gain a deeper understanding of the structural composition of the MAZ@Zn coating, X-ray diffraction (XRD) patterns of bare Zn and MAZ@Zn samples were recorded, as shown in [Figure 1M](#). In addition to the diffraction peaks similar to those of Zn (PDF#04-0813), the MAZ@Zn sample displays additional peaks that align with the characteristics of MoS₂ (PDF#37-1492)^[57]. This observation confirms that the MAZ@Zn coating has been successfully applied to the Zn foil. Furthermore, the diffraction peaks of M7AZ3@Zn, MAZ@Zn, and M3AZ7@Zn are similar. FTIR analyses provide additional verification of the presence of Alg-Zn in the coatings, as illustrated in [Supplementary Figure 1B](#). The peak at 3,112 cm⁻¹ is indicative of the O-H stretching vibration in the hydrogen bonds of the alginate salt polymer chain, the peak around 1,593 cm⁻¹ corresponds to the asymmetric stretching vibrations of the -COO- group on the alginate salt chain, and the peak near 1,011 cm⁻¹ is likely attributed to the C-O stretching vibration^[43].

XRD analyses of coated samples and bare Zn after a five-day immersion in a 2 M ZnSO₄ solution were conducted, to investigate the stability of different Zn anodes [[Supplementary Figure 2](#)]. For the bare Zn electrode, besides the diffraction peaks of Zn (PDF#04-0831), two peaks appear at 16.25° and 24.47°, which correspond to irreversible formation of Zn₄(SO₄)₄(OH)₆·5H₂O (PDF#39-0688), indicating that severe passivation occurred during deposition^[58]. In contrast, for M7AZ3@Zn, MAZ@Zn and M3AZ7@Zn electrodes, no trace of the by-product is observed. These observations highlight the stability of the MAZ coating in ZnSO₄ electrolytes and its ability to shield the Zn anode from direct contact with the electrolyte, which effectively inhibits potential side reactions. In [[Supplementary Figure 3](#)], EIS shows that the impedances of M7AZ3@Zn, MAZ@Zn and M3AZ7@Zn are lower than those of bare Zn. Compared with M7AZ3@Zn and M3AZ7@Zn, MAZ@Zn exhibits the lowest impedance, whereas M3AZ7@Zn shows the highest impedance. After soaking in the electrolyte for 8 h, the impedance of M7AZ3@Zn, M3AZ7@Zn, MAZ@Zn and bare Zn increases, while the impedance of MAZ@Zn changes least before and after resting^[32].

In assessing the electroplating/stripping stability of bare Zn, M7AZ3@Zn, MAZ@Zn, and M3AZ7@Zn, the electrochemical behaviors of the electrodes in symmetric cells at 1 mA cm⁻² and 1 mA h cm⁻² were recorded. As shown in [Figure 2A](#), the bare Zn-symmetric cell shows poor cycle performance, experiencing short-circuit problems in less than 200 h of testing. Specifically, symmetric cells assembled with M7AZ3@Zn and M3AZ7@Zn demonstrate extended cycling lifetimes of 560 h and 770 h, respectively. Notably, the symmetric cell composed of MAZ@Zn electrodes achieves an impressive operational duration of 880 h while maintaining low polarization. As shown in [Supplementary Figure 4A](#), M7AZ3@Zn, MZA@Zn, M3AZ7@Zn and bare Zn have polarization values of 59, 56, 57 and 62 mV, respectively^[46]. The overpotential is calculated by the difference between the tip potential and the subsequent stabilization potential, which is closely related to the kinetics of the Zn stripping/plating reaction^[59]. The lowest polarization is observed for MAZ@Zn; this is consistent with the EIS results [[Supplementary Figure 3](#)]. Such excellent cycling stability and low polarization can be attributed to the optimized coating composition and interfacial structure. The Alg-Zn matrix offers ionic pathways, while uniformly dispersed MoS₂ nanosheets guide Zn²⁺ deposition and suppress side reactions. With an appropriate MoS₂ content, the MAZ@ coating achieves a balanced structure that synergistically enhances interfacial stability and electrochemical performance. Overall, the MAZ coating enables uniform Zn deposition and superior cycling stability among the modified anodes.

Further charge-discharge test was executed with a higher current density of 4 mA cm⁻² (1 mAh cm⁻²), as shown in [Figure 2B](#). The results reveal that bare Zn encounters a short circuit after approximately 200 h [[Figure 2B](#)]. In sharp contrast, MAZ@Zn exhibits a cycle life of more than 500 h, with a nucleation overpotential of 107 mV compared to 137 mV for bare Zn, showing a considerable reduction in polarization [[Supplementary Figure 4B](#)]. These results finally confirm that MAZ@Zn coating significantly enhances the stability of Zn stripping and plating processes. Compared with most previously reported interfacial modification strategies [[Supplementary Table 1](#)], the MAZ@Zn anode exhibits superior cyclic stability and cumulative capacity, although there is still room for further improvement in overall electrochemical performance.

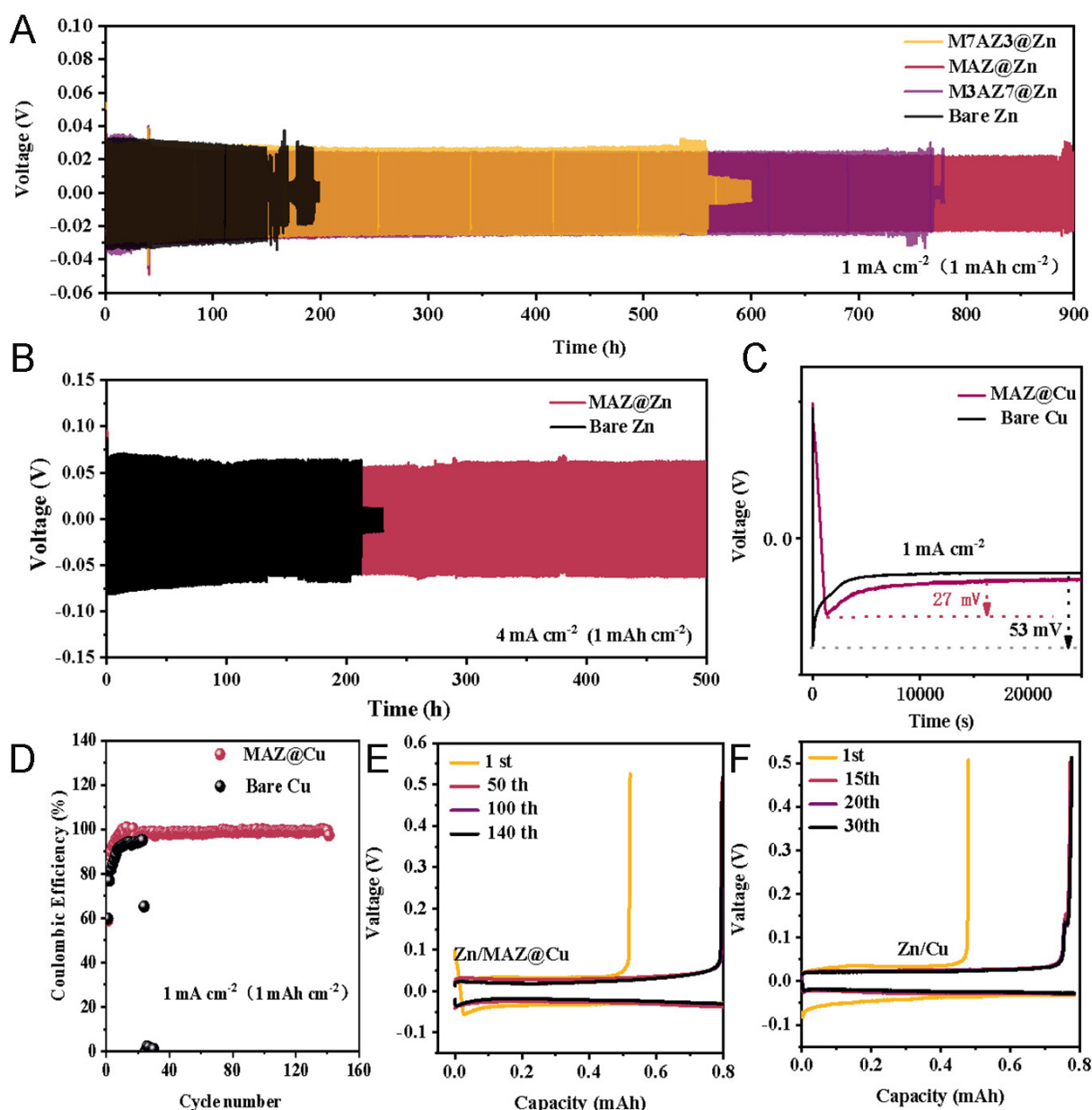


Figure 2. Electrochemical performance in Zn/Cu cells and Zn/Zn cells. Constant current charge and discharge curves of symmetric cells of bare Zn, M7AZ3@Zn, MAZ@Zn and M3AZ7@Zn: (A) at 1 mA cm⁻² (1 mAh cm⁻²); (B) 4 mA cm⁻² (1 mAh cm⁻²). (C) Nucleation overpotential in Zn/Cu and Zn/MAZ@Cu in asymmetric cells. (D) CE of Zn plating/stripping in Zn/Cu and Zn/MAZ@Cu half-cells. GCD curves of (E) Zn/MAZ@Cu and (F) Zn/Cu half-cells.

Under a discharge current density of 1 mA cm⁻² [Figure 2C], the measured nucleation overpotential of Zn/Cu is 53 mV. In contrast, for Zn/MAZ@Cu, the nucleation overpotential is significantly lower, measured at 27 mV, which is smaller than that of the bare Zn. To better understand the effect of the MAZ coating on the Zn electroplating/stripping process, we measured the Coulombic efficiency (CE) of Zn/Cu and Zn/MAZ@Cu half-cells at a current density of 1 mA cm⁻² and an area-specific capacity of 1 mAh cm⁻². As depicted in Figure 2D-F, the Zn/Cu half-cell exhibits voltage fluctuations after only 30 cycles due to the formation of Zn dendrites, which can lead to internal short circuits. In contrast, the Zn/MAZ@Cu half-cell maintains stable CE even after 140 cycles, demonstrating excellent reversibility in Zn stripping and electroplating. This result not only underscores the efficacy of the MAZ coating in preserving the electrochemical reversibility of Zn, but also signifies its role in effectively mitigating side reactions and curtailing dendritic growth of Zn.

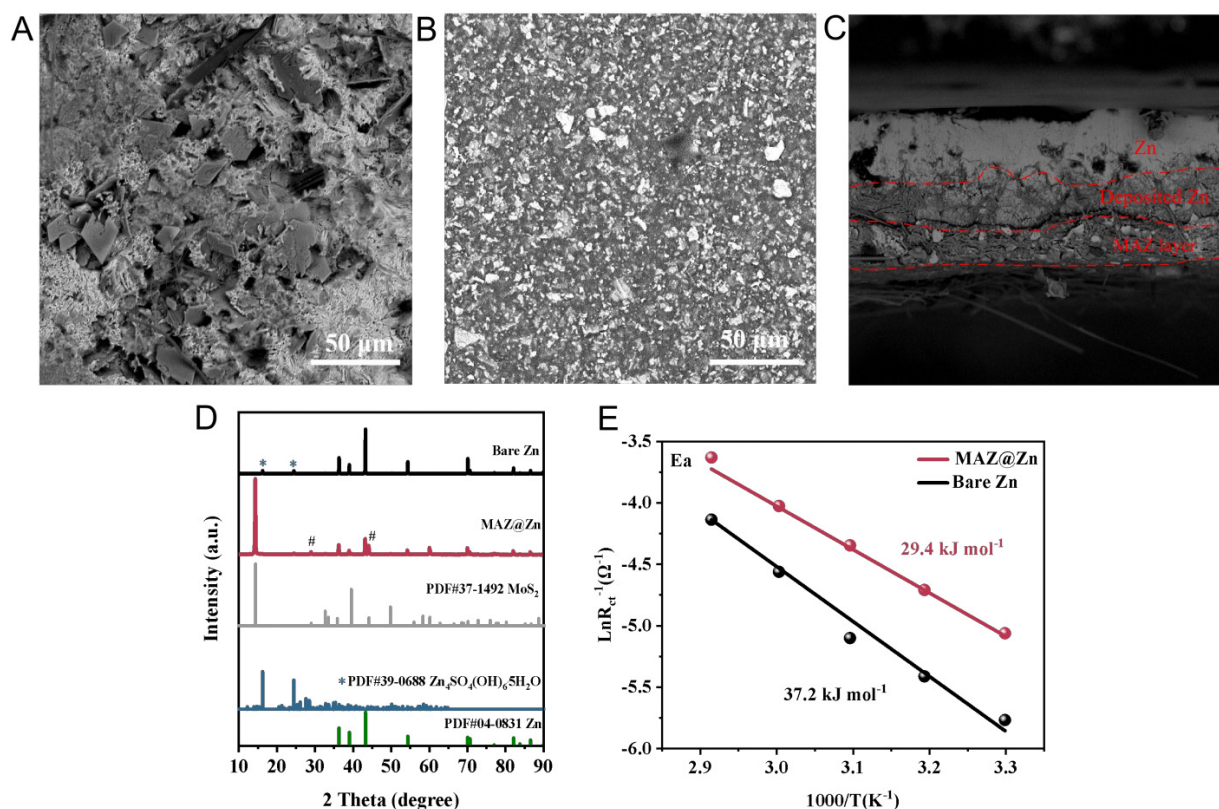


Figure 3. Structural and electrochemical analysis of Zn-based cells. SEM images of (A) bare Zn, (B) the top surface, and (C) the cross-section of MAZ@Zn in symmetric cells after 50 cycles at 1 mA cm^{-2} (1 mAh cm^{-2}). (D) XRD patterns of bare Zn and MAZ@Zn in symmetric cells after 50 cycles at 1 mA cm^{-2} (1 mAh cm^{-2}); (E) Corresponding Arrhenius plots and comparison of activation energies between bare Zn and MAZ@Zn.

To further validate the ion-transport characteristics of the Alg-Zn/MoS₂ hybrid coating, EIS was conducted to evaluate its intrinsic ionic conductivity. Nyquist plots were recorded at open-circuit potential over 100 kHz – 0.01 Hz using stainless steel (SS)||SS and MAZ@SS||MAZ@SS symmetric cells with a glass fiber separator [Supplementary Figure 5]. The MAZ coating exhibited an ionic conductivity of approximately $4 \times 10^{-2} \text{ S cm}^{-1}$, which is higher than that of the previously reported Alg-Zn coating ($1.83 \times 10^{-2} \text{ S cm}^{-1}$), indicating its superior ion-transport capability. Furthermore, to provide direct evidence of Zn²⁺-preferential transport, the Zn-ion transference number ($t_+(\text{Zn}^{2+})$) was determined by chronoamperometry combined with EIS following the Bruce-Vincent method [Supplementary Figure 6]. The Zn symmetric cell (without coating) shows $t_+(\text{Zn}^{2+}) = 0.42$, whereas the MAZ@Zn symmetric cell shows $t_+(\text{Zn}^{2+}) = 0.52$, indicating more preferential Zn²⁺ migration through the Alg-Zn framework in the presence of the MAZ coating. These results provide direct experimental evidence for the fast and selective ion-transport capability of the MAZ coating, which contributes to the enhanced electrochemical performance of the MAZ@Zn anode.

SEM and XRD investigations were conducted for the bare Zn and MAZ@Zn after cycling in symmetric cells. These cells underwent 50 cycles at a current density of 1 mA cm^{-2} and area-specific capacity of 1 mAh cm^{-2} . The bare Zn electrode displays a rough and uneven surface, characterized by a dense array of dendritic structures and non-uniform deposition of Zn [Figure 3A]^[60]. In sharp contrast, the surface of the MAZ@Zn electrode remains smooth and uniform, with no signs of Zn dendrite formation or cluster aggregates [Figure 3B]. The cross-sectional image [Figure 3C] further confirms that the MAZ coating remains intact, with Zn deposition occurring uniformly on the coating surface, forming a dense and compact layer. XRD analysis provided further evidence of the protective role of MAZ coating. It shows the presence of the by-product $\text{Zn}_4\text{SO}_4(\text{OH})_6 \cdot 5\text{H}_2\text{O}$ (PDF#39-0688) is observed in the bare Zn samples, but no trace of the by-product is found in the MAZ@Zn electrode [Figure 3D].

In addition, when the MAZ coating was removed from the cycled MAZ@Zn electrode for XRD analysis [Supplementary Figure 7], only the diffraction peak of MoS₂ (PDF#37-1492) was observed in the coating material, while no peak of Zn (PDF#04-0831) was detected. This indicates that, under the regulation of the MAZ coating, Zn is uniformly and effectively deposited beneath the coating at the interface between the coating and the Zn substrate, which is consistent with the cross-sectional results shown in Figure 3C. Furthermore, to further elucidate the deposition behavior, a single Zn deposition test was conducted using a MAZ@Cu substrate [Supplementary Figure 8]. The use of Cu foil eliminates the influence of the Zn base electrode, enabling direct observation of Zn deposition morphology. Both the cross-sectional and top-view SEM images (before and after coating removal) confirm that Zn is deposited beneath the coating, forming a uniform and compact interfacial layer. Temperature-dependent EIS measurements [Supplementary Figure 9] and the corresponding Arrhenius analysis [Figure 3E] revealed a lower desolvation activation energy for MAZ@Zn symmetric cells (29.4 kJ mol⁻¹) compared with bare Zn cells (37.2 kJ mol⁻¹). This reduced energy barrier indicates that the MAZ coating enables more efficient Zn²⁺ desolvation and accelerates interfacial charge transfer.

In-situ optical microscopy was used for real-time observation of the Zn electroplating and stripping processes. As shown in Supplementary Figure 10, under a current density of 0.3 mA cm⁻², bare Zn displays an uneven surface Zn deposition even after applying the current for 15 min. In contrast, under the guiding effect of the MAZ coating, the MAZ@Zn electrode demonstrates a much more uniform deposition at the same time interval. The difference becomes even more pronounced after 60 min. The surface of the bare Zn electrode is covered by extensive and irregular Zn deposits. On the other hand, the MAZ@Zn electrode exhibits a remarkably uniform deposition. The ability of the MAZ coating to promote uniform deposition of Zn is a key factor in enhancing the longevity and efficiency of Zn electrodes for AZIBs.

To further evaluate the suppression of side reactions, Tafel and linear sweep voltammetry (LSV) measurements were carried out [Supplementary Figure 11]. The LSV curves reveal that the MAZ@Zn electrode exhibits a higher HER overpotential, while the Tafel plots show a lower corrosion current density compared with bare Zn, confirming that the coating effectively inhibits both HER and corrosion. These findings further validate the proposed “ion-sieve” function of MoS₂: while the low migration energy barrier facilitates Zn²⁺ transport, the barrier effect against H₂O helps to suppress parasitic reactions, thereby enhancing the interfacial stability of the Zn anode.

Ab initio DFT calculations were performed to investigate the adsorption behavior of Zn atoms on both 2H-MoS₂ and Zn surfaces. Figure 4A shows the lattice model of the 2H-MoS₂(002) surface, which corresponds to the most intense XRD peak in Figure 1I and possesses superior chemical stability^[61,62]. A supercell consisting of two MoS₂ layers with a 40 Å vacuum in the z-direction to mimic the 2H-MoS₂ (002) surface. Five possible Zn adsorption sites were examined on the 2H-MoS₂ surface, as illustrated in Figure 4B: atop a Mo atom (site 1), atop a S atom (site 2), nestled between two Mo atoms (site 3), between two S atoms (site 4), and at the nexus of three Mo atoms (site 5). As a benchmark, the adsorption of Zn atoms on a Zn (002) surface was also studied [Figure 4C]^[63]. The Zn (002) facet was selected because it is the most thermodynamically stable plane of hexagonal Zn, exhibiting the lowest surface energy among the exposed facets^[64–66]. Therefore, it is widely used as a benchmark surface for modeling Zn adsorption and diffusion behavior in both theoretical and experimental studies. The Zn layers are categorized as either the “A” or “B” layer based on their geometric configurations, represented by gray and black spheres in Figure 4B. To systematically explore Zn adsorption, three typical sites were considered on the Zn (002) surface: directly above a Zn atom from the A layer (between-Zn), directly above a Zn atom from the B layer (above-B-Zn), and interstitially between two surface Zn atoms (above-A-Zn). The calculated adsorption energies are -81.5, -106.9, and -111.4 kJ mol⁻¹ for the above-A-Zn, above-B-Zn, and between-Zn sites, respectively, as presented in Figure 4D. Notably, the between-Zn site exhibits the least exothermic adsorption energy, indicating that Zn adsorption at this

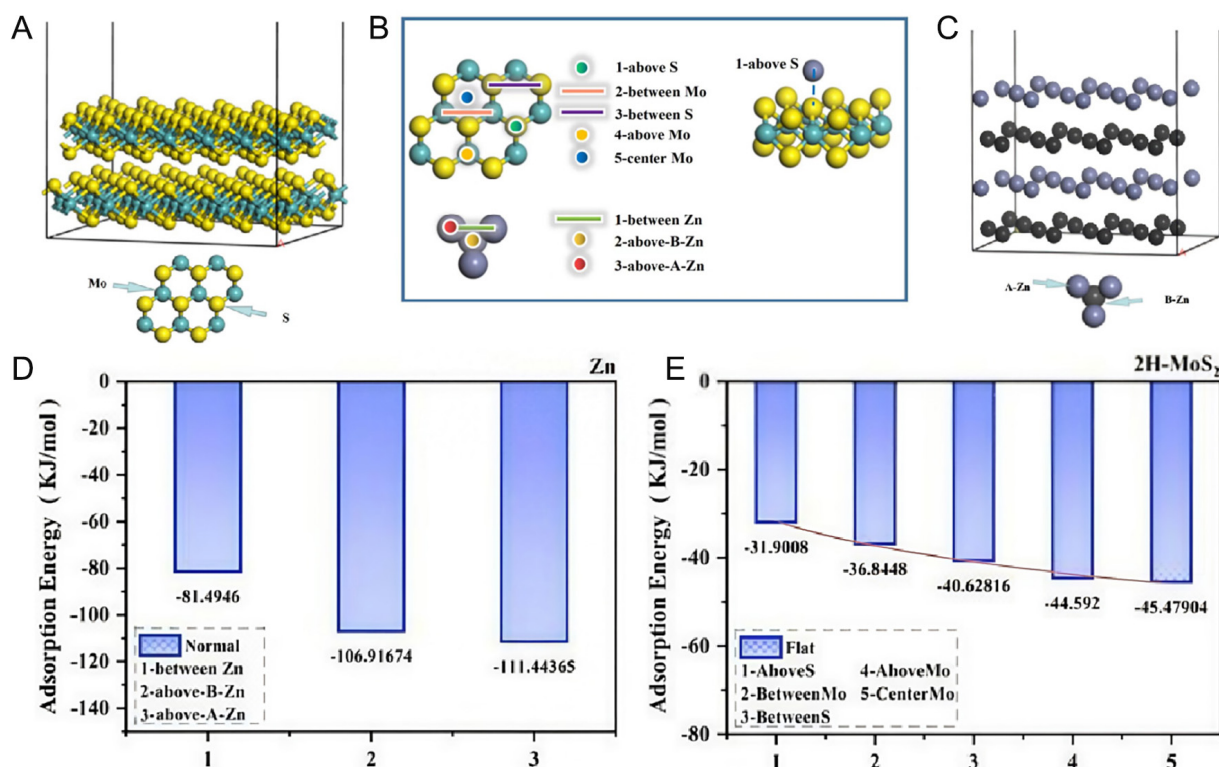


Figure 4. Lattice structure and adsorption of Zn atoms on 2H-MoS₂ and Zn surfaces. Lattice structure of 2H-MoS₂ (002) surface (A) and with Mo and S atoms plotted as green and yellow spheres, respectively. (B) Selected adsorption sites on the MoS₂ (002) and Zn (002) surfaces. (C) Lattice structure of Zn (002) surface, with Zn atoms plotted as gray(black) spheres. (D) Adsorption energy of Zn atom at different sites of Zn (002) surface and (E) 2H-MoS₂ (002) surface.

position perturbs the intrinsic hexagonal close-packed (HCP) structure of the Zn surface due to its interstitial configuration. In contrast, the above-A-Zn and above-B-Zn sites show comparable, more negative adsorption energies, suggesting a small diffusion barrier for Zn migration between these two energetically favorable sites. As summarized in Figure 4E, the adsorption energies of Zn atoms on the 2H-MoS₂ (002) surface range from -31.9 to -45.5 kJ mol⁻¹, which are significantly weaker than those on the Zn (002) surface. Combined with the smaller energy differences among the adsorption sites, this suggests a reduced diffusion energy barrier for Zn migration on 2H-MoS₂, enabling homogeneous Zn distribution, suppressed dendritic growth, and improved interfacial stability.

In addition to the theoretical analysis of Zn adsorption on the MoS₂ surface, we further examined whether Zn²⁺ ions could also be intercalated into the MoS₂ interlayers. Interlayer insertion of Zn²⁺ is strongly dependent on the MoS₂ phase (1T/2H), interlayer chemistry/spacing, defect characteristics, and the testing protocol^[67–71]. In this study, commercial, pristine 2H-MoS₂ was used as a surface-modifying layer without further modification, and limited Zn-storage activity is therefore expected. To substantiate this, pure MoS₂ electrodes were evaluated in 2 M ZnSO₄. CV and GCD measurements [Supplementary Figure 12] show nearly no reversible capacity within the selected potential window, indicating no measurable Zn²⁺ intercalation into the S-Mo-S galleries. This observation is consistent with reports that pristine 2H-MoS₂ requires defect engineering to effectively accommodate Zn²⁺^[69]. Accordingly, in this study, MoS₂ primarily regulates Zn²⁺ adsorption and guides uniform Zn deposition, rather than serving as a Zn²⁺ storage host.

To comprehensively evaluate the performance of MAZ@Zn in full cells, NVO was employed as the cathode material. As shown in Supplementary Figure 13, the NVO displays a nanowire-like morphology, and its crystalline phase matches well with PDF#16-0601. The electrolyte consisted of a mixed solution of 2 M ZnSO

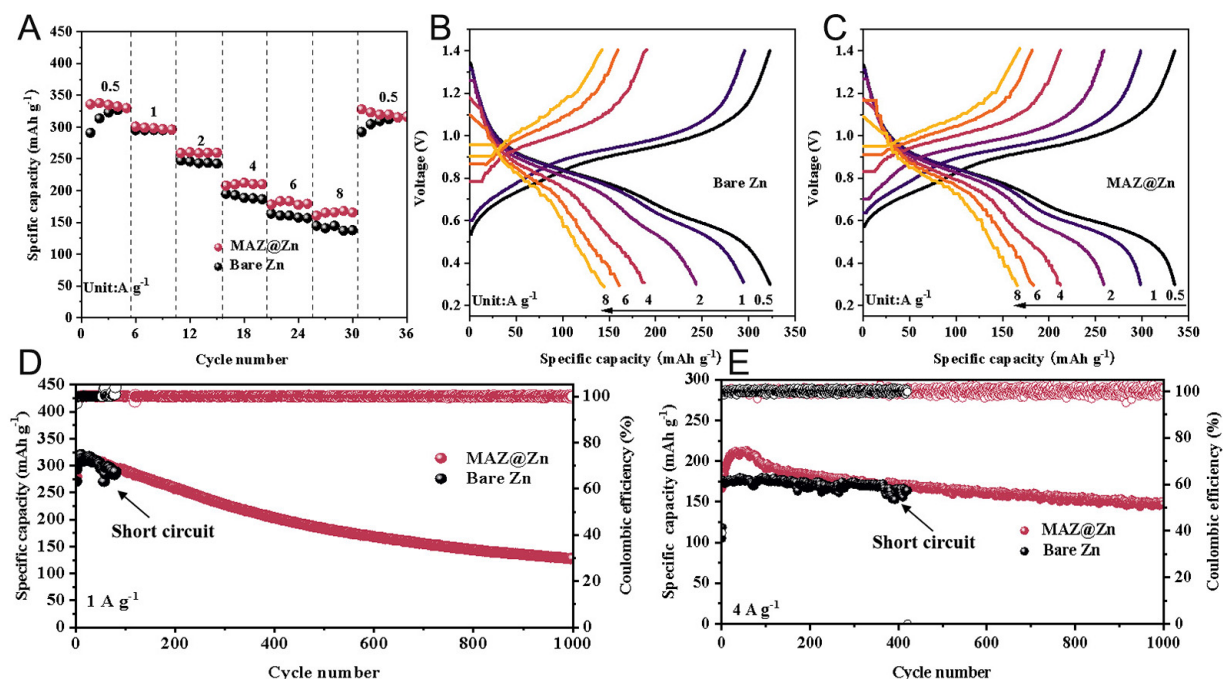


Figure 5. Comparison of electrochemical performance between bare Zn//NVO and MAZ@Zn//NVO batteries. (A) Rate performance (charge-discharge curves) at a current density from 0.5 A g⁻¹ to 8 A g⁻¹; GCD curves of (B) bare Zn//NVO; (C) MAZ@Zn//NVO; (D) cycling test at 1 A g⁻¹; (E) cycling test at 4 A g⁻¹.

and 1 M Na₂SO₄. Bare Zn and MAZ@Zn were used as the anode electrodes, respectively. Preliminary CV tests were conducted at a scan rate of 0.1 mV s⁻¹, with a working voltage window set at 0.3–1.4 V. The results of the second cycle test, as shown in [Supplementary Figure 14](#), indicate that in the NVO full battery, whether MAZ@Zn or bare Zn is used as the anode, the CV curve exhibits two pairs of redox peaks. Specifically, the redox peaks for bare Zn are at 0.59/0.77 V and 0.83/1.0 V, while those for MAZ@Zn are at 0.58/0.78 V and 0.82/1.02 V, which are similar to those of bare Zn^[72]. The similarity of the electrochemical reaction processes between MAZ@Zn and bare Zn anodes in NVO full batteries was further confirmed. The electrochemical performance of Zn//NVO and MAZ@Zn//NVO batteries at different current densities was further evaluated [Figure 5A]. At current densities of 0.5, 1, 2, 4, 6, and 8 A g⁻¹, the discharge specific capacities of the MAZ@Zn//NVO full battery are 332.8, 302.3, 262.8, 215.2, 183.6, and 166.3 mAh g⁻¹, respectively, higher than those of the bare Zn//NVO battery, which are 322.9, 295.3, 243.4, 188.7, 160.7 and 144.7 mAh g⁻¹ at corresponding current densities. When the battery returns to a current density of 0.5 A g⁻¹ from 8 A g⁻¹, the MAZ@Zn//NVO battery shows an excellent capacity retention rate. The results suggest that MAZ@Zn//NVO exhibits better rate performance in comparison to Zn//NVO, which is attributed to the enhanced charge transfer process, as indicated by the EIS results [Supplementary Figure 15]. Figure 5B and C shows the constant current charge and discharge curves of the two batteries from 0.5 A g⁻¹ to 8 A g⁻¹. The results are consistent with the CV curve, with two obvious voltage platforms and the average working voltage reaching 0.83 V (vs. Zn²⁺/Zn). In addition, MAZ@Zn//NVO shows a superior cycle performance compared with bare Zn//NVO at 1 A g⁻¹ and 4 A g⁻¹ [Figure 5D and E]. For MAZ@Zn//NVO, the specific discharge capacity remains 130.6 mAh g⁻¹ at 1 A g⁻¹ over 1,000 cycles with a capacity retention of 45.7%. For bare Zn//NVO, it can only be cycled for 80 times. At 4 A g⁻¹, the discharge capacity of MAZ@Zn//NVO is 147 mAh g⁻¹ over 1,000 cycles, and the capacity retention is 88.4%, while for bare Zn//NVO, it failed at 420 cycles. The results surpass most previously reported results for AZIBs with vanadium oxide-based cathodes [Supplementary Table 2]. The excellent cycle and rate performance of MAZ@Zn//NVO is attributed to the improved interface properties of Zn and the high diffusion rate of Zn²⁺ on the MoS₂ surface endowed by the MAZ coating, enabling fast charge transfer and uniform Zn deposition during cycling.

CONCLUSIONS

In conclusion, facile MAZ@Zn hybrid coating composed of Alg-Zn and MoS₂ is effective in suppressing Zn dendrite formation and enhancing the cycle stability of Zn anodes for AZIBs. The Alg-Zn guides the Zn²⁺ ion diffusion in the coating; in addition, first-principles calculations reveal that the 2H-MoS₂ (002) surface significantly reduces the migration energy barriers for Zn²⁺, functioning as an efficient ion sieve that promotes fast migration and uniform distribution of Zn²⁺ ions across the electrode surface. The hybrid coating effectively suppresses the formation of Zn dendrites and lowers the charge transfer energy of the Zn anode. The MAZ@Zn anode demonstrates remarkable electrochemical performance. In symmetric cells, the MAZ@Zn anode achieves a cycle life of 880 h at a current density of 1 mA cm⁻² (1 mAh cm⁻²), significantly outperforming bare Zn electrodes. The high ionic conductivity and Zn²⁺ transference number of the Alg-Zn/MoS₂ coating further confirm its capability to facilitate rapid and uniform Zn²⁺ transport at the electrode interface, which contributes to the improved cycling stability. Moreover, the MAZ coating effectively suppresses the HER by impeding H₂O access to the Zn surface, further improving interfacial stability and prolonging the cycle life of Zn anodes. In full Zn//NVO batteries, the MAZ@Zn anode maintains a discharge capacity of 130.6 mAh g⁻¹ after 1,000 cycles at 1 A g⁻¹, and a capacity retention rate of 88.4% after 1,000 cycles at 4 A g⁻¹. For the bare Zn anode, the full battery cycles only 80 times at 1 A g⁻¹ and 420 times at 4 A g⁻¹. The MAZ hybrid coating offers a practical and effective solution to improve the performance of Zn anodes for high-performance and long-life AZIBs.

DECLARATIONS

Acknowledgements

The authors wish to thank Changzhou Jiulian Battery Materials Co., Ltd. for providing the fiberglass separators used in this study.

Authors' contributions

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Availability of data and materials

[Supplementary Material](#) is available from the authors.

Financial support and sponsorship

This work was financially supported by the financial support from the National Natural Science Foundation of China (52122209, 12174270, 52131306, 52073143 and 52403001).

Conflicts of interest

Prof. Wu, Y. is Editor-in-Chief of *Energy Materials*. He was not involved in any part of the editorial process, including reviewer selection, manuscript handling, or decision-making, while the other authors have declared that they have no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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Supplementary Materials

[Supplementary Materials](#)

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