

Challenges and Strategies for Zinc Metal Anode

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The primary aim of this concept is to point the overlooked critical issues, and summary the crucial roles of regulating the electric double layer (EDL), zinc ion transfer number and electrolyte design, thereby enhancing the stability of zinc metal anodes and expediting the commercialization of aqueous zinc-ion batteries. Aqueous zinc-ion batteries (AZIBs) are received intensive attention for their inherent safety, economic feasibility, competitive electrochemical performance, and environmental friendliness. However, the rampant side reactions such as dendrite growth, hydrogen evolution, corrosion, and passivation of zinc metal anode seriously hinder the stability of AZIBs. This concept is aimed to enhancing the practical viability of

AZIBs, with a particular focus on the stability of zinc metal anodes. Here, we summarized the strategies for achieving high reversibility zinc metal anode, and points out the shortcomings that still exist in current research. Then the crucial role of regulation electric double layer (EDL), zinc ion transfer number and electrolyte design is summarized in detail. Finally, we consider that the design of zinc anode should focus more on practicality and commercialization, and provide a perspective for the future development direction of zinc anode. We hope to provide an inspiration for the design of stable zinc metal anodes in AZIBs from this concept.

Introduction

Aqueous zinc ion batteries (AZIBs) are one of the candidates for the next generation of safe rechargeable batteries, which have attracted widespread attention in the fields of energy storage devices.^[1–3] However, the development of AZIBs has been plagued by dendrite growth, hydrogen evolution reaction, corrosion and passivation in zinc metal anode.^[4,5] In the past few decades, researches on zinc metal anode have mainly focused on electrolyte design,^[6] membrane modification,^[7,8] interface engineering^[9–12] and non-liquid solid electrolyte^[13] (Figure 1). These strategies successfully regulated the uniform deposition of Zn²⁺ to a certain extent, effectively improving the stability of anode/electrolyte interface. Thus, it is greatly inhibited the uncontrollable dendrites and harmful side reactions, achieving a long life-span AZIBs.^[14,15]

Nonetheless, the journey towards commercial viability remains a significant challenge. A multitude of critical concerns necessitates immediate and focused research efforts in the realm of zinc anode development: *i)* Unstable anode/electrolyte interface. The non-uniform distribution of ions, pH fluctuations during cycling, and the formation of irregular by-products at zinc anode interface, contribute to unstable anode/electrolyte interface. The unstable anode/electrolyte interface will further exacerbate dendrite formation and side reactions, severely impacting cycling stability. *ii)* Neglected ion transfer numbers. The transfer number is an important parameter that affects the stability of anode/electrolyte interface. A low zinc ion transfer number ($t_{\text{Zn}^{2+}}$) leads to the concentration gradient near the zinc anode due to low conductivity, which triggers dendrite growth

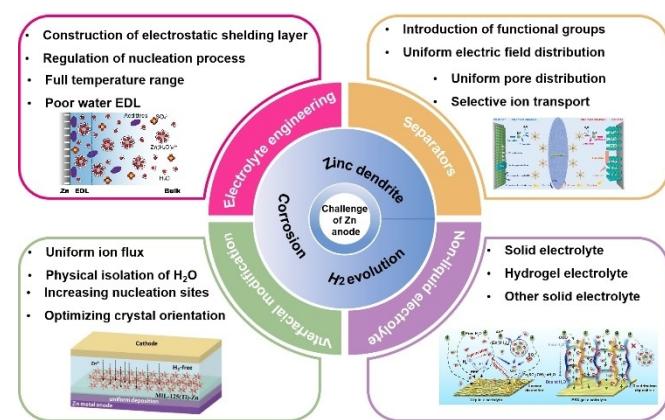


Figure 1. Challenges and strategies of zinc metal anode: electrolyte engineering^[6] (Reproduced with permission from Copyright 2024 Wiley), separators^[7] (Reproduced with permission from Copyright 2023 Wiley), interfacial modification^[9] (Reproduced with permission from Copyright 2022 Elsevier) and non-liquid electrolyte^[13] (Reproduced with permission from Copyright 2022 Elsevier).

and anion-related side reactions. *iii)* Optimal electrolyte selection for enhanced performance. The significant thermodynamic instability of zinc anode in aqueous electrolyte will spontaneous corrosion, resulting in uneven interface. Electrolyte engineering can effectively reduce the corrosion of free water molecules, thereby improving the stability of the zinc anode. To address these key issues and expedite the commercialization process of AZIBs, this concept emphasizes the strategies to overcome these major challenges and proposes the next research direction and prospects for zinc metal anode.

The Anode/Electrolyte Interface

The stability of AZIBs is primarily reliant on the characteristics of the anode/electrolyte interface.^[16] As shown in Figure 2, the

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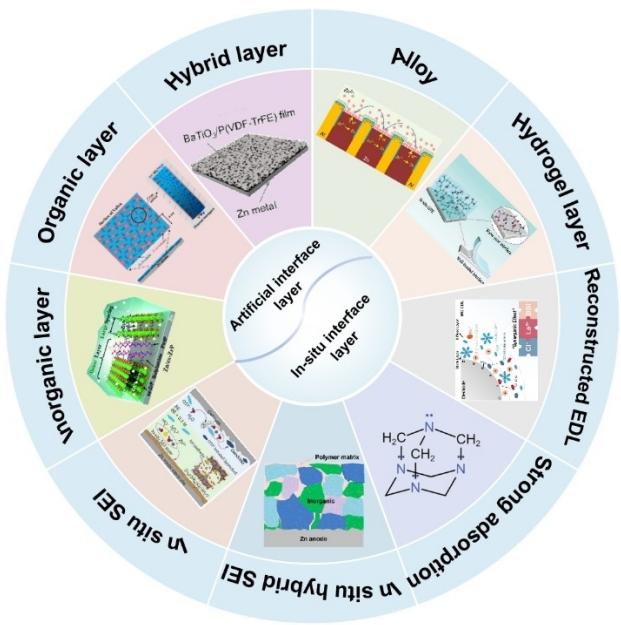


Figure 2. Research progress on zinc metal electrode interface: Artificial interface layer (inorganic layer, organic layer, hybrid layer and alloy)^[17–20] (Reproduced with permission from Copyright 2021 Royal Society of Chemistry; Copyright 2020, Wiley; Copyright 2023 American Chemical Society; Copyright 2020 Springer) and in-situ interface layer (hydrogel layer Copyright 2022 Wiley) reconstructed EDL (Copyright 2024 Wiley) strong adsorption (Copyright 2023 Wiley), in-situ hybrid SEI (Copyright 2023 Wiley), in-situ SEI (Copyright 2022 Elsevier).^[21–25]

main strategies to construct a stable anode/electrolyte interface in current include constructing artificial interface layers and forming in situ protective interfaces.^[17–25] Compared to ex-situ artificial protective coatings, in-situ protective interfaces exhibit the advantages such as remarkable adhesion of membrane with

substrate, uniform and stable coating composition, and enhanced ion conductivity.^[26] Up to now, extensive researches have been carried out on the in-situ formation of SEI on the surface of zinc through electrolyte additives. The performance of the battery affected by the thickness and composition of the SEI. Nevertheless, it is impossible to precisely control the thickness of SEI due to the intricate electrochemical environment during the formation process. Moreover, SEI might undergo alterations during multiple charge and discharge cycles. Particularly at high voltages or high current densities, SEI could rupture, peel off, or regenerate. This may result in an increase in the internal resistance of the battery, capacity degradation, and even have impact on the safety of the battery.^[27,28] Consequently, it is essential to seek more efficacious methods to enhance the stability of the anode/electrolyte interface.

EDL consists of dense layer and diffusion layer, and the dense layer can be further divided into inner Helmholtz plane (IHP) and outer Helmholtz plane (OHP).^[29] In general, the polar H₂O molecules tend to occupy the IHP of zinc anode in ZnSO₄ electrolyte, causing the hydrogen evolution reaction (HER) an inevitable occurrence.^[30] During HER, the OH⁻ will accumulate on the interface forming insulating and uneven Zn₄SO₄(OH)₆·xH₂O by-products, thereby influencing the coulombic efficiency (CE) and reducing the stability of the AZIBs. In OHP, the hydrated [Zn(H₂O)₆]²⁺ diffuses from the diffusion layer to the OHP for desolvation, and then under the action of electrostatic force, the desolvated Zn²⁺ in the OHP will be reduced and deposited on the electrode.^[31] Specifically, the high desolvation energy of [Zn(H₂O)₆]²⁺ in the OHP will increase local polarization, giving rise to an increase in overpotential and a higher risk of HER.^[32] Moreover, the desolvation energy dominates the charge/discharge kinetics of AZIBs, especially under high rate conditions. Additionally, the high desolvation



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energy will impede the transfer of zinc ions between the OHP and the IHP, causing an uneven zinc ion flux resulting in dendrite formation. Therefore, the synergistic regulation of the H₂O content in the IHP and the desolvation process of [Zn(H₂O)₆]²⁺ in the OHP is the most effective approach to suppressing dendrite growth and interface parasitic side reactions.

Recently, Xu et al. introduced the trace zwitterion additive into AZIBs (Aqueous Zinc-Ion Batteries) to construct a multi-functional self-adsorption interface.^[33] The TFA⁻ anions, which possess strong specific adsorption capabilities, can adsorb on the surface of the zinc metal anode during the entire plating/stripping process. This adsorption effectively reduces the H₂O content within the IHP, thereby suppressing side reactions such as hydrogen evolution and corrosion. Meanwhile, the Py⁺ located in the OHP can enhance the desolvation process of [Zn(H₂O)₆]²⁺, thus optimizing the reaction kinetics of AZIBs. Owing to the synergistic modulation of IHP and OHP, the zinc anode is capable of stable plating/stripping for 10,000 hours. Moreover, the Zn||I₂ full cell demonstrates an outstanding cycle stability of 30,000 cycles. This work presents an effective approach to protecting the zinc metal anode by regulating IHP and OHP, which represents a novel contribution in the current research field.

The Zinc Ion Transfer Number

The ion transfer number also possesses an influence on the stability of the anode/electrolyte interface. An electrolyte with a high zinc ion transfer number is capable of effectively diminishing concentration polarization, restraining the formation of zinc dendrites, and enhancing the electrochemical performance of the battery.^[34] Nevertheless, in the prior

strategies for modulating the zinc metal anode, the significant characteristic of the zinc ion transfer number of the electrolyte was neglected.

During the charging/discharging process, the electric field existing between the cathode and anode drives the transfer of anions and cations within the electrolyte. For instance, in the commonly used ZnSO₄ electrolyte, under the influence of the electric field directed towards the anode, the positively charged Zn²⁺ ions will migrate towards the anode, whereas anions such as SO₄²⁻ will migrate towards the cathode. However, in reality, only the migration of the working ions like Zn²⁺ is of significance for AZIBs. In liquid electrolytes, Zn²⁺ can become solvated due to the solvation effect, which restricts the desolvation process and the migration of Zn²⁺. In contrast, anions are scarcely solvated. This implies that the majority of ion conductivity is mainly contributed by anions. Consequently, the ion transfer number of Zn²⁺ in liquid electrolytes is typically less than 0.5, and the effective conductivity of the electrolyte is substantially diminished.^[35] Such a phenomenon will result in the accumulation of anions on the surface of the cathode electrode, creating a concentration gradient between the anode and cathode. This, in turn, gives rise to concentration polarization and induces a large overpotential, thereby limiting the energy density and power density of AZIBs.

As illustrated in Figure 3, we have summarized the representative works in the past three years on strategies to increase the zinc ion transfer number.^[36-45] Currently, there are three primary methods for improving the transfer number of zinc ions: (i) Enhancing the desolvation process of Zn²⁺. By applying molecular sieve coatings or similar treatments to the zinc metal anode's surface, the desolvation of [Zn(H₂O)₆]²⁺ is accelerated. This reduction in the desolvation energy barrier subsequently promotes the migration of zinc ions. (ii) Establishing a dedicated ion channel to enhance Zn²⁺ transport. By

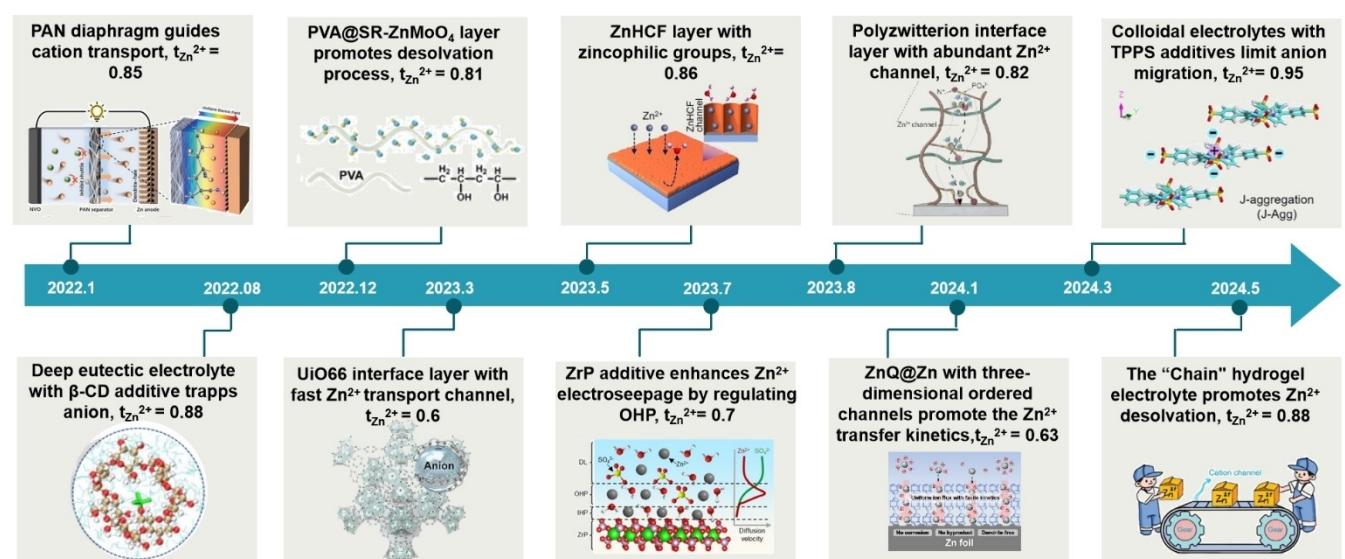


Figure 3. Strategies for increasing the transfer number of zinc ions in AZIBs in representative works over the past three years.^[36-45] (Reproduced with permission from Copyright 2021 Wiley; Copyright 2022 Wiley; Copyright 2023 Royal Society of Chemistry; Copyright 2023 Springer; Copyright 2023 Wiley; Copyright 2023 Wiley; Copyright 2023 Wiley; Copyright 2024 Wiley; Copyright 2024 Wiley).

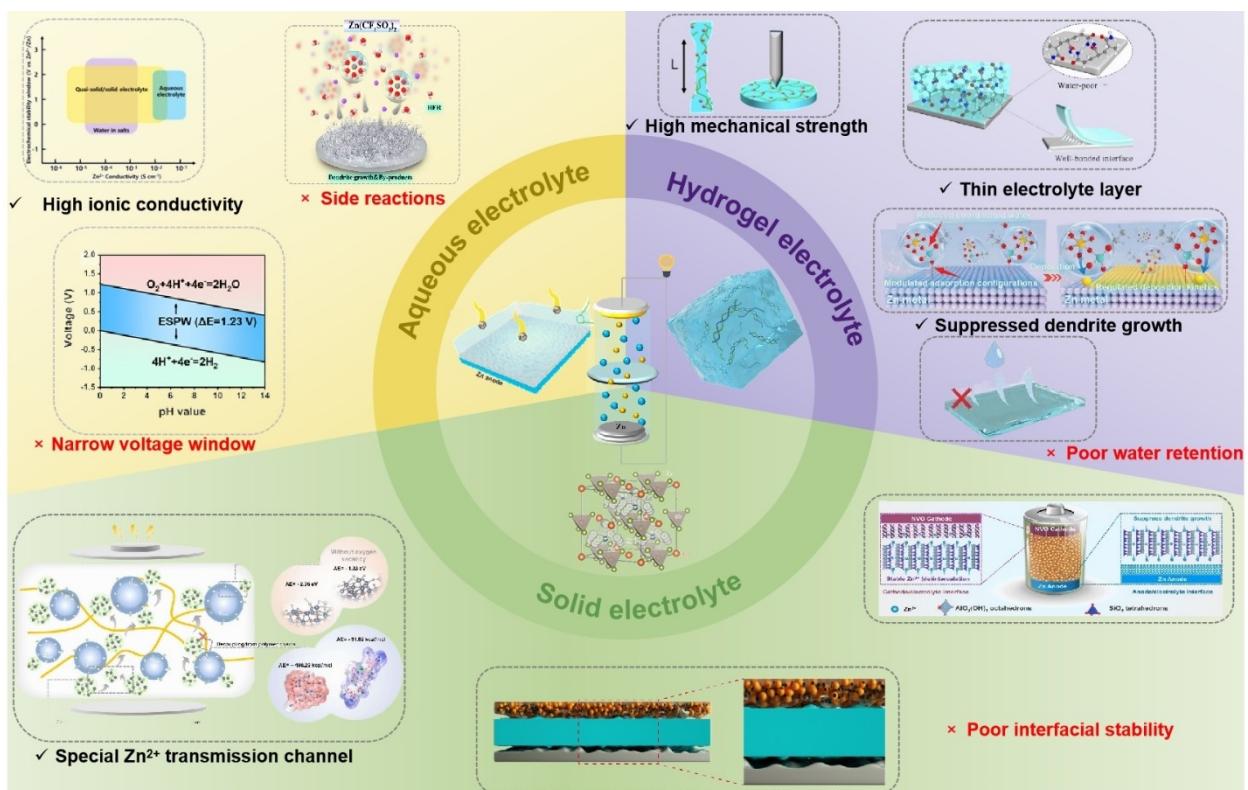


Figure 4. The advantages and disadvantages of aqueous electrolyte^[46–48] (Reproduced with permission from Copyright 2022 Wiley; Copyright 2022 Elsevier; Copyright 2022 Springer Nature), hydrogel electrolyte^[49–51] (Reproduced with permission from Copyright 2024 American Chemical Society; Copyright 2023 Elsevier; Copyright 2024 Wiley; Copyright 2022 Wiley) and solid electrolyte^[52–54] ((Reproduced with permission from Copyright 2024 Wiley; Copyright 2024 Wiley; Copyright 2024 Wiley).

repelling negatively charged anions and attracting positively charged cations, a swift transport pathway for Zn^{2+} is created, thereby promoting the transfer of zinc ions. (iii) Reducing free anions through anion binding strategies. By anchoring anions via covalent bonds or anion receptors, the transfer number of anions is minimized, ensuring that Zn^{2+} is the dominant transfer ion in the electrolyte, thus increasing the transfer number of zinc ions. In a word, future research should give more consideration in improving the zinc ion transfer number.

The Design Criteria for Electrolytes

In addition to the anode/electrolyte interface and zinc ion transfer number, electrolyte also plays a pivotal role in the reaction mechanisms and kinetics within batteries. Electrolytes are broadly categorized into three types: liquid, quasi-solid (hydrogel) and solid electrolytes, as depicted in Figure 4. Liquid electrolytes possess more superior ionic conductivity,^[46–48] yet they face persistent issues such as unstable electrode/electrolyte interfaces, hydrogen evolution side reactions, and the challenges of electrolyte volatilization and leakage, which have hindered the advancement of aqueous zinc-ion batteries (AZIBs). Hydrogel electrolytes, rich in hydrophilic groups, can modulate the solvation structure around zinc ions and restrict the amount of free H_2O molecules, mitigating detrimental

effects on the zinc metal anode.^[49–51] Nevertheless, side reactions originate from residual free H_2O molecules are still inevitable.

The solid state electrolytes have the characteristics of excellent thermal stability, high potential window and environmentally friendly, making them an ideal choice to replace traditional electrolytes.^[52–54] While solid-state lithium-ion batteries have received extensive research interest in recent years, the application of solid-state electrolytes in zinc batteries remains nascent. Unlike lithium-ion, the mobility of zinc ions in solid-state electrolytes is significantly hindered by strong electrostatic interactions, presenting greater challenges in the engineering of solid-state electrolytes for zinc-ion batteries.^[55] Moreover, the advancement of solid-state zinc batteries need to address the intrinsic limitations of solid electrolytes: *i*) The relatively low ionic conductivity. The ionic conductivity of solid-state electrolytes is generally inferior than their liquid electrolytes, which constrains the reaction kinetics and rate capabilities of the batteries. To enhance conductivity, further refinement in the material composition and structural design of solid electrolytes is imperative. *ii*) The inferior interface compatibility. The inadequate interfacial contact between solid-state electrolytes and electrodes poses a significant challenge for solid-state batteries. Such interfaces can lead to increased resistance, adversely impacting the performance and cycle life of the batteries.

Despite the inherent limitations of solid electrolytes, their compelling benefits make them as a pivotal avenue for advancing battery technology. Shi et al. have reported a zinc-based solid electrolyte using the layered natural mineral kaolin (KL Zn).^[53] This KL Zn solid electrolyte expand the voltage window to 2.73 V. The ion conductivity reaches 5.08 mScm^{-1} , and zinc ion transfer number elevates to 0.79. Besides, Yang et al. have engineered a solid-state Zn-I₂ battery employing the inorganic ZnP₃ (ZPS) solid electrolyte, effectively addressing the depressing interfacial challenges that plague aqueous electrolytes.^[56] The inorganic ZnP₃ electrolyte exhibits a low Zn²⁺ diffusion energy barrier of about 0.3 eV, excellent ion conductivity of $2.0 \times 10^{-3} \text{ Scm}^{-1}$ (30°C), high chemical/electrochemical stability, and mechanical strength.

The aforementioned strategies have primarily concentrated on improving ion conductivity of solid electrolyte. At present, the research on solid electrolytes in zinc batteries is rarely reported. It can get some effective strategies from the lithium-ion batteries to enhance the compatibility at the solid electrolyte-zinc anode interface. For instance, Yu et al. successfully constructed an integrated cathode/polymer electrolyte for solid state LMBs (Li//DICE-LFP//DSICE) through polymer chain exchange and recombination of dynamic disulfide bonds and supramolecular hydrogen bonds in the molecule. This design fundamentally enhances the interfacial contact between the solid electrolyte and the electrode, ensuring a more continuous lithium ion (Li⁺) transport pathway at the cathode-electrolyte interface. It also contributes to the stabilization of uniform Li⁺ deposition on the lithium anode surface throughout the cycling process.^[57] Ilias Belharouak et al. applied short duration high-voltage pulses to poorly formed interfaces, which can significantly reduce contact impedance.^[58] The localized joule heating effect induced by these pulses results in a high local current density near the interface pores, which can significantly enhance the contact between lithium and the solid electrolyte (LALZO).

Solid state electrolytes are expected to bring revolutionary changes to the field of batteries, offering safer, more efficient, and high stability battery solutions. Future researches will focus on material innovation, interface optimization, and manufacturing process improvement to promote the commercialization of solid-state zinc batteries. The development of solid electrolytes will provide strong support for the utilization of sustainable energy and the advancement of electronic devices.

Summary and Outlook

This concept discusses the existing problems and current researches of zinc metal anode, and elaborates on several key issues that need to be addressed in detail, including unstable electrode/electrolyte interfaces, neglected Zn²⁺ ion transfer number, and the design of solid electrolytes. Although some progress has been made in the research of zinc metal anode, as shown in Figure 5, future researches should still focus on practicality and commercial value, explore advanced character-

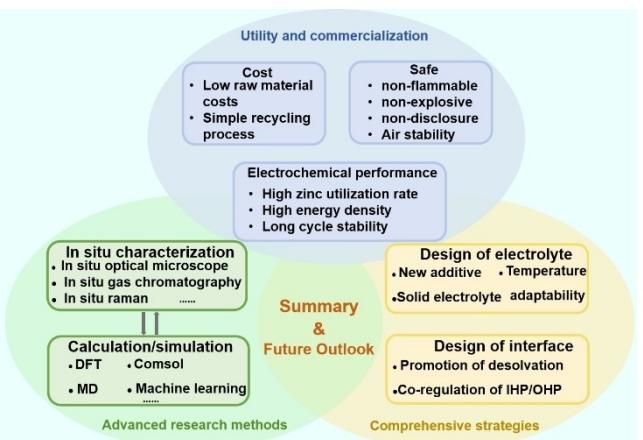


Figure 5. Summary and future outlook of zinc anode in AZIBs.

ization and simulation methods, and develop more comprehensive strategies.

- Considering on practicality and commercialization.** Considering the future commercialization of AZIBs, it is crucial to prioritize the development and utilization of low-cost materials and technologies suitable for industrialization. Although many studies have demonstrated the technical capability to suppress dendrite formation and side reactions, the high cost of materials and complex processes involved make their commercial application challenging. Some of these approaches, however, increase the cost of AZIBs, contradicting their inherent low-cost advantage. Therefore, for potential commercial applications, it is important to seek new methods that are both cost-effective and high-performance. When designing a stable anode/electrolyte interface, it is essential to balance the simplicity of the chosen method on the cost of the selected electrolyte additives. Solution replacement method is an effective method to prepare zinc anode artificial interface, which has the characteristics of high efficiency, variety and economic feasibility. When using electrolyte additives to reshape zinc anode interface, the economic applicability of the additives should be considered. In terms of solid electrolyte design, polymer electrolyte has the characteristics of low cost and easy processing, which is the preferred ideal choice for solid electrolyte materials. In addition, facile and economical methods should be chosen in the design of strategies to increase the number of zinc ion migrations. In addition, in previous studies, excess zinc was usually present in the form of thick zinc foils ($\geq 100 \mu\text{m}$ in thickness). Excessive zinc constantly supplements active zinc to overcome losses caused by "dead zinc" and by-products, resulting in deceptive long cycle life and high coulombic efficiency. Moreover, the excessive zinc metal increases the cost of the battery and reduces the energy density of full battery. Therefore, more effective strategies must be adopted to suppress the growth of zinc dendrites and reduce the formation of by-products, improving zinc utilization efficiency, and thus increasing the energy density of AZIBs.

- ii) **Exploring advanced characterization and simulation methods.** The more advanced analysis and monitoring methods are needed to analyze the evolution process of various components of the zinc anode during charging and discharging. Combining simulation calculation with experimental results to enhance the understanding of the mechanisms of anode failure processes, thereby facilitating the discovery of effective solutions to these issues. DFT calculation, MD simulation, and COMSOL simulation are three common methods employed in current research on zinc metal anodes. Machine learning will be an effective tool for AZIB design, which can improve the accuracy of predicting battery performance, pointing potential research direction.
- iii) **Developing more comprehensive strategies.** The challenges associated with zinc anodes are interconnected. Thus, there is a pressing need to seek strategies that delivers multifunctional benefits in a single, cohesive approach. For example, some electrolyte additives can not only disrupt the solvation structure by coordinating with Zn^{2+} , but also adsorb onto metal surfaces to intercept the corrosion of H_2O molecules. Stable anode interface can be achieved by combining structural anode with additives or introducing additives into gel electrolyte.

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Conflict of Interests

The authors declare no conflict of interest.

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