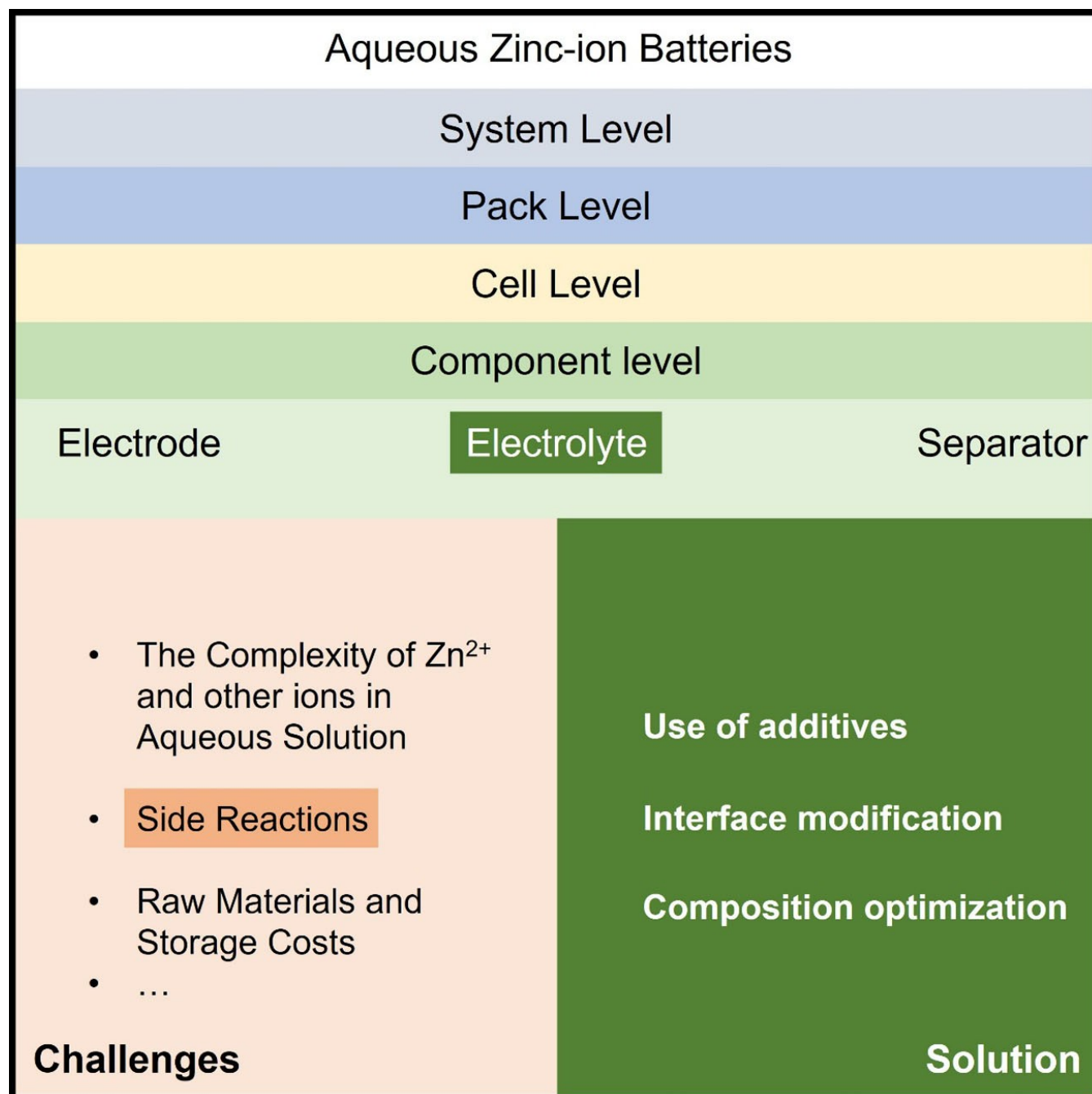


Recent Advances of Aqueous Electrolytes for Zinc-Ion Batteries to Mitigate Side Reactions: A Review

Xuan Gao,^[a, b] Haobo Dong,^{*[a]} Claire J. Carmalt,^[a] and Guanjie He^{*[a, b]}



The paper discusses the challenges associated with the performance of zinc-ion batteries (ZIBs), such as side reactions that lead to reduced capacity and lifespan. The strategies for mitigating side reactions in ZIBs, including additives, electrolyte-electrode interface modification, and electrolyte composition optimization, are explored. Combinations of these approaches may be necessary to achieve the best performance for ZIBs. However, continued research is needed to improve the commercial viability of ZIBs. Areas of research requiring

attention include the understanding of the mechanisms behind side reactions in ZIBs and the development of cost-effective and scalable manufacturing processes for ZIBs with available electrolyte. By developing effective strategies for mitigating side reactions, researchers can improve the efficiency and lifespan of ZIBs, making them more competitive with lithium-ion batteries in various applications, including grid energy storage.

1. Introduction

Zinc-ion batteries (ZIBs) are a type of rechargeable battery that utilize zinc ions as the charge carrier. The development of ZIBs is gaining attention due to their potential for low-cost and high-performance energy storage systems.^[1] Zinc is abundant, inexpensive, and environmentally friendly, making it an attractive material for energy storage applications. One critical component of ZIBs is the electrolyte, which plays a crucial role in the performance and stability of the battery.^[2] The electrolyte is a liquid or gel substance that facilitates the movement of ions between the anode and cathode of the battery during charging and discharging. It also helps to maintain the chemical stability of the electrodes and prevent unwanted side reactions that can degrade the battery's performance.^[3]

The choice of electrolyte in ZIBs is crucial, as it can significantly impact the battery's energy density, power output, and cycle life.^[4] Commonly used electrolytes for ZIBs include aqueous and non-aqueous solutions of zinc salts such as zinc sulfate, zinc chloride, zinc triflate, etc.^[5] Aqueous electrolytes are attractive due to their low cost, high conductivity, and safety. Despite the potential advantages of aqueous electrolyte, there are several challenges that must be addressed to enable their widespread use. One of the most significant challenges is the formation of side reactions during cycling, which can lead to the degradation of the battery's performance and shorten its lifespan.^[6]

Side reactions can occur when the electrolyte reacts with the electrodes or other components of the battery, leading to the formation of unwanted products that can interfere with the battery's function. For example, when using aqueous electrolytes, the formation of hydrogen gas during charging can lead to gas evolution and reduced battery performance.^[7] Similarly,

the formation of zinc dendrites on the anode during cycling can cause short circuits and damage to the battery.^[8] To address these challenges, researchers are investigating new electrolytes that can improve the performance and stability of ZIBs. Some effective concepts, such as "water in salt", pH buffer, and ion solvation structure improvement have been carried out in ZIBs electrolytes, and some good progress has been made.^[9]

In this mini review, we will discuss the different types of side reactions that can occur in ZIBs electrolytes and their impact on battery performance and safety. Then, some post-2021 research progress on these side effects will also be summarized. This paper is mainly to provide valuable reference to researchers for future research in ZIBs electrolyte.

2. Side reactions in Electrolytes of ZIBs

Like all aqueous electrochemical systems, aqueous ZIBs are subject to various side reactions that can impact their performance and safety.

2.1. Water decomposition

In aqueous media, the decomposition of water usually involves two half-reactions of oxygen evolution reaction (OER) and hydrogen evolution reaction (HER), which require four and two electron transfers respectively, and the reaction pathway is related to the pH value of the electrolyte and the structure of the electrode surface, while H₂/O₂ evolution. A suitable electrolyte solvent should have a wide electrochemical window (EW). The most widely used and simplest strategy is to use the energy level difference between the lowest unoccupied molecular orbital (LUMO) and the highest occupied molecular orbital (HOMO) to represent the electrolyte EW. This estimation based on LUMO and HOMO usually causes an error in overestimating the EW. Researchers are also using more methods to obtain better estimates, such as EW prediction methods based on thermodynamic cycles.^[10] Water has an inherent thermodynamic oxidation potential (oxygen evolution reaction, OER) and reduction potential (hydrogen evolution reaction, HER) with a voltage window of 1.23 V between them, and its narrow EW limits the operating voltage, resulting in a lower energy density, as shown in Figure 1. Additionally, the formation of hydroxide ions can lead to the degradation of the electrolyte and the formation of by-products on the electrode surface, which can

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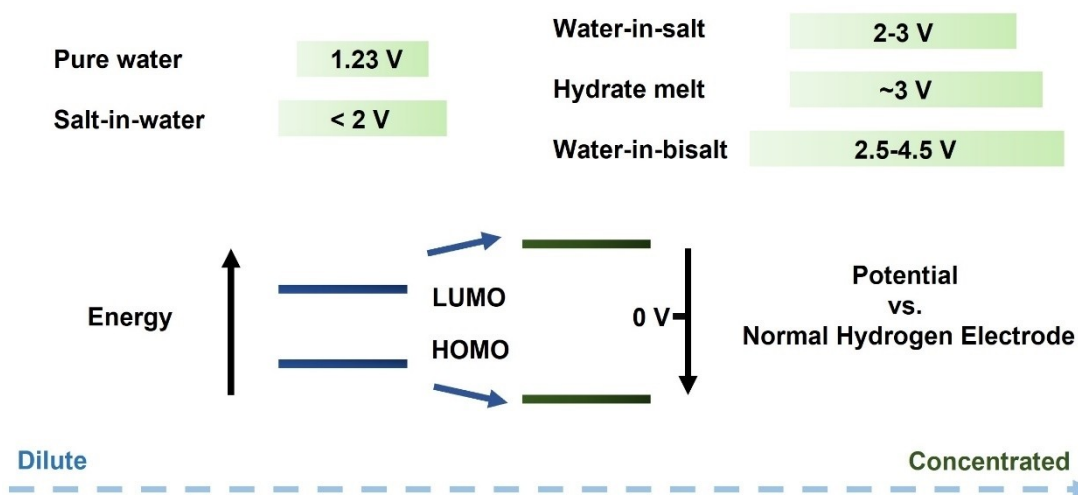


Figure 1. Illustration of output voltage extension from the standpoint of overpotential using water-in-salt or hydrate-melt electrolytes. Traditional aqueous electrolyte is referred to as the salt-in-water electrolyte.

further reduce battery performance.^[11] The HER/OER occurring at the electrode surface consumes a portion of the electrons that should be provided to the active material, resulting in low Coulombic efficiency, cell swelling, and continuous consumption of electrolyte. Therefore, suppressing water splitting and broadening the operating voltage window are crucial for the development of aqueous batteries. Strategies of water-in-salt, pH control, isotope effects, SEI modification, electrolyte additives, artificial protective layers, polymer coatings, ionic liquids, etc.^[12] have been used to broaden the stable EW, and further stabilize and expand the output voltage of aqueous batteries.^[13]

2.2. Zinc dendrites

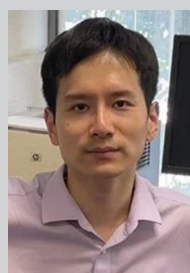
Zinc dendrite formation is another side reaction that can occur in ZIBs.^[14] Zinc metal is relatively abundant and can be directly used as a negative electrode, which is one of the reasons why aqueous ZIBs have attracted much attention. But repeated deposition/stripping can also cause uneven deposition, resulting in dendrites. Dendrites are small, needle-like structures that form on the surface of the zinc anode during battery operation.^[15] These structures can pierce the separator and cause a short circuit, leading to reduced battery performance and safety.^[16] The strategies of inhibiting zinc dendrites has the following aspects: (1) adding inert components to the aqueous electrolyte solution; (2) protective layer on the anode; (3) 3D



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porous electrode structure design to reduce the local current or; (4) new separator, etc.

2.3. Corrosion (self-discharge)

Side reactions such as corrosion and passivation while charging and discharging have a strong relationship with Coulombic efficiency and cycle stability.^[17] Corrosion, or self-discharge, is a side reaction that occurs in strong acid or strong alkaline aqueous solutions, which not only affects the Coulombic efficiency but also causes irreversible water consumption and electrolyte concentration changes, often at the electrode-electrolyte interface. Hence, limiting the chemical interaction at the electrode-electrolyte interface is essential for preventing electrode corrosion. Addition of inorganic or organic corrosion inhibitors to electrodes or electrolytes is the most often described strategy for reducing the corrosion rate of electrode materials at this time.

3. Mitigation strategies for side reactions

The impact of side reactions on battery performance and safety can be significant.^[18] As mentioned before, HER and OER both consume electrons that could otherwise be used to charge the battery, leading to reduced battery performance. Additionally, the formation of hydrogen gas and hydroxide ions can cause the battery to swell and rupture, posing a safety risk. To mitigate the impact of these side reactions, various strategies have been developed. One approach involves the use of additives in the electrolyte to suppress water decomposition and dendrite formation.^[19] For example, the addition of a small amount of an organic solvent to the electrolyte can suppress hydrogen evolution by reducing the availability of protons at the electrode, such as diethylenetriamine, glycine, etc.^[11,20] Similarly, the addition of organic and inorganic salt to the electrolyte can suppress water decomposition by reducing the availability of water molecules at the electrode as well, such as $\text{Co}(\text{CH}_3\text{COO})_2$, NiSO_4 , etc.^[5,21] However, the use of additives can also introduce new problems, such as decreased battery performance or increased cost.^[22] Moreover, the effect of additives can depend on the concentration. Another approach involves the optimization of the electrolyte-electrode interface and electrolyte composition. For example, the use of a three-dimensional zinc anode can reduce the formation of dendrites by providing a larger surface area for the deposition of zinc ions.^[23]

3.1. Use of additives

Additives can improve the stability of the electrolyte by suppressing unwanted side reactions or by forming a passivation layer on the electrode surface.^[24] For example, organic additives such as sorbitol have been used to suppress the side effect in the electrolyte. Some pH stabilizers can effectively

alleviate the uneven deposition of zinc on the anodes. In 2023, He et al. introduced $\text{NH}_4\text{H}_2\text{PO}_4$ (NHP) to stabilize the pH value during ZIBs cycling and suppress dendrite generation and side reactions, as shown in Figure 2.^[12b] NHP helps regulate Zinc deposition and suppress side reactions, thereby improving battery performance. The NH_4^+ form a protective layer on the surface of the Zinc, preventing direct contact with water. The combination of NH_4^+ and H_2PO_4^- maintains a favorable pH at the electrode-electrolyte interface, which enable highly reversible galvanization and stripping process in ZIBs.

3.2. Modification of the electrolyte-electrode interface

The interface between the electrolyte and the electrode is critical for the performance and stability of the battery. The modification of the interface can prevent or reduce the side reactions and improve the efficiency of the charge-discharge cycles.^[25] One approach is the use of protective coatings on the electrode surface, such as carbon coatings, metal oxide coatings, or polymer coatings.^[26] These coatings can improve the adhesion between the electrode and the electrolyte, reducing the formation of cracks or gaps that can lead to unwanted reactions. The improvement of the interface can also be achieved by modification of electrolyte.^[27] Gel polymer electrolytes (GPEs) have been demonstrated to inhibit the formation of zinc dendrites and ameliorate parasitic reactions by limiting ion migration/diffusion and water content. Wang et al. presented a chemical welding approach including the in-situ creation of a gel electrolyte that allows ZIBs to attain ultralong life and reversibility.^[28] Gel electrolyte is generated spontaneously on the zinc anode surface by redox polymerization initiated by zinc metal. The direct inclusion of the zinc anode in the chemical production of the gel electrolyte results in a well-bonded and water-poor interface between the electrode and electrolyte, which not only reduces side reactions but also facilitates preferential (002) zinc deposition. The resulting in-situ symmetric cell has an extremely long lifetime of 5100 h and a hybrid capacitor including the in-situ electrolyte operates smoothly for over 40,000 cycles at 20 A g^{-1} , as shown in Figure 3.

3.3. Optimization of the electrolyte composition

The composition of the electrolyte can affect the performance and stability of the battery by controlling the ion transport, the chemical reactions, and the thermal properties.^[29] The optimization of the electrolyte composition can involve several parameters, such as the choice of solvent, the concentration of salt, the pH, and the presence of additives. The addition of salts such as $\text{Zn}(\text{TFSI})_2$ can improve the conductivity and the stability of the electrolyte by increasing the dissociation of zinc ions.^[30] In 2023, Gao et al. introduced the electrochemical effect of isotopes (EEI) of water into the electrolyte of ZIBs to solve the problem of severe side reactions and massive gas generation.^[12a] Due to the low diffusion coefficient and high coordination of ions in D_2O , the possibility of side reactions is reduced, resulting in a larger

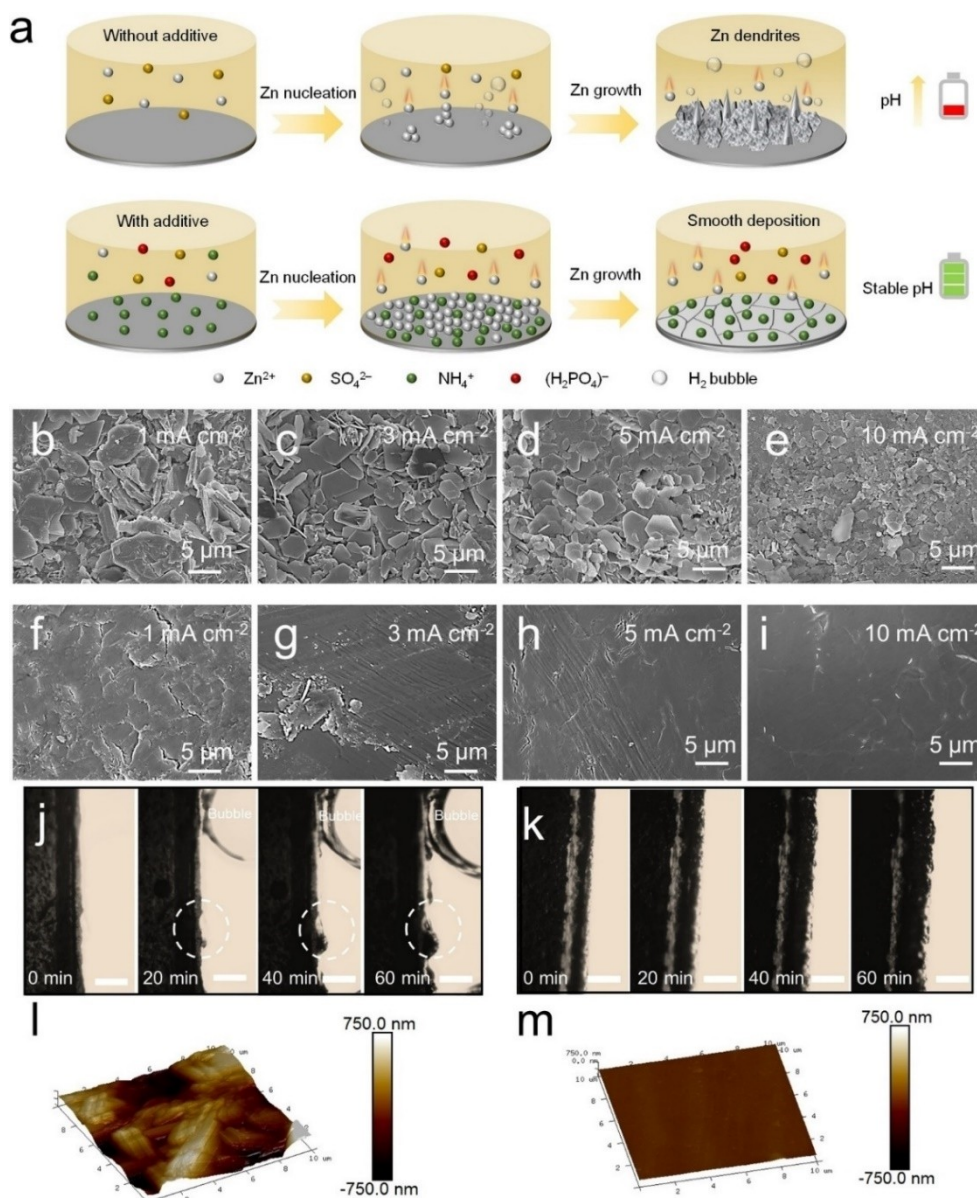


Figure 2. Plating/stripping morphologies of zinc. (a) A schematic depicting the impact of NHP additions on the zinc deposition process. Zinc deposits on a zinc substrate (Zn | Zn symmetric cells) with current densities from 1 mA cm⁻² to 10 mA cm⁻² with 1 mAh cm⁻² in (b–e) 1 M ZnSO₄ (BE) and (f–i) 1 M ZnSO₄ + 25 mM NHP (DE). In situ optical microscope pictures of zinc plating on a zinc substrate at 10 mA cm⁻² in j) 1 M ZnSO₄ (BE) and k) 1 M ZnSO₄ + 25 mM NHP (DE) with a scale bar of 300 μm. Atomic force microscopy (AFM) images of zinc electrodes that have been cycled in l) 1 M ZnSO₄ (BE) and m) 1 M ZnSO₄ + 25 mM NHP (DE).^[12b] Reproduced from ref. [12b] Copyright (2023), with the permission from Wiley-VCH.

EW, less pH change, and less zinc hydroxide sulfate (ZHS) generation. Furthermore, D₂O can remove various ZHS phases induced by changes in bound water during cycling, thereby forming a stable interface between the electrode and the electrolyte. Notably, the D₂O-based electrolyte exhibits stable wide EW cycle performance (Figures 4a–d) and less gas generation (Figure 4f). Full cells using D₂O-based electrolytes, as shown in Figures 4e and g, were shown to be more stable after 1,000 cycles over a wide voltage window of 0.8–2.0 V and 3,000 cycles over a typical voltage window of 0.8–1.9 V at a current density of 2 Ag⁻¹ cyclic behavior, exhibiting ~100% reversible efficiency.

4. Conclusions

ZIBs have garnered significant interest in recent years due to their low cost, high safety, and abundant resource availability. However, the performance of ZIBs has been limited by the occurrence of side reactions, such as water decomposition, zinc dendrites and corrosion, that occur during the electrochemical processes of the batteries. These side reactions cause irreversible changes to the electrodes, resulting in reduced capacity and shortened lifespan. As a result, it is crucial to develop effective strategies for mitigating side reactions in ZIBs by additives, electrolyte-electrode interface modification and elec-

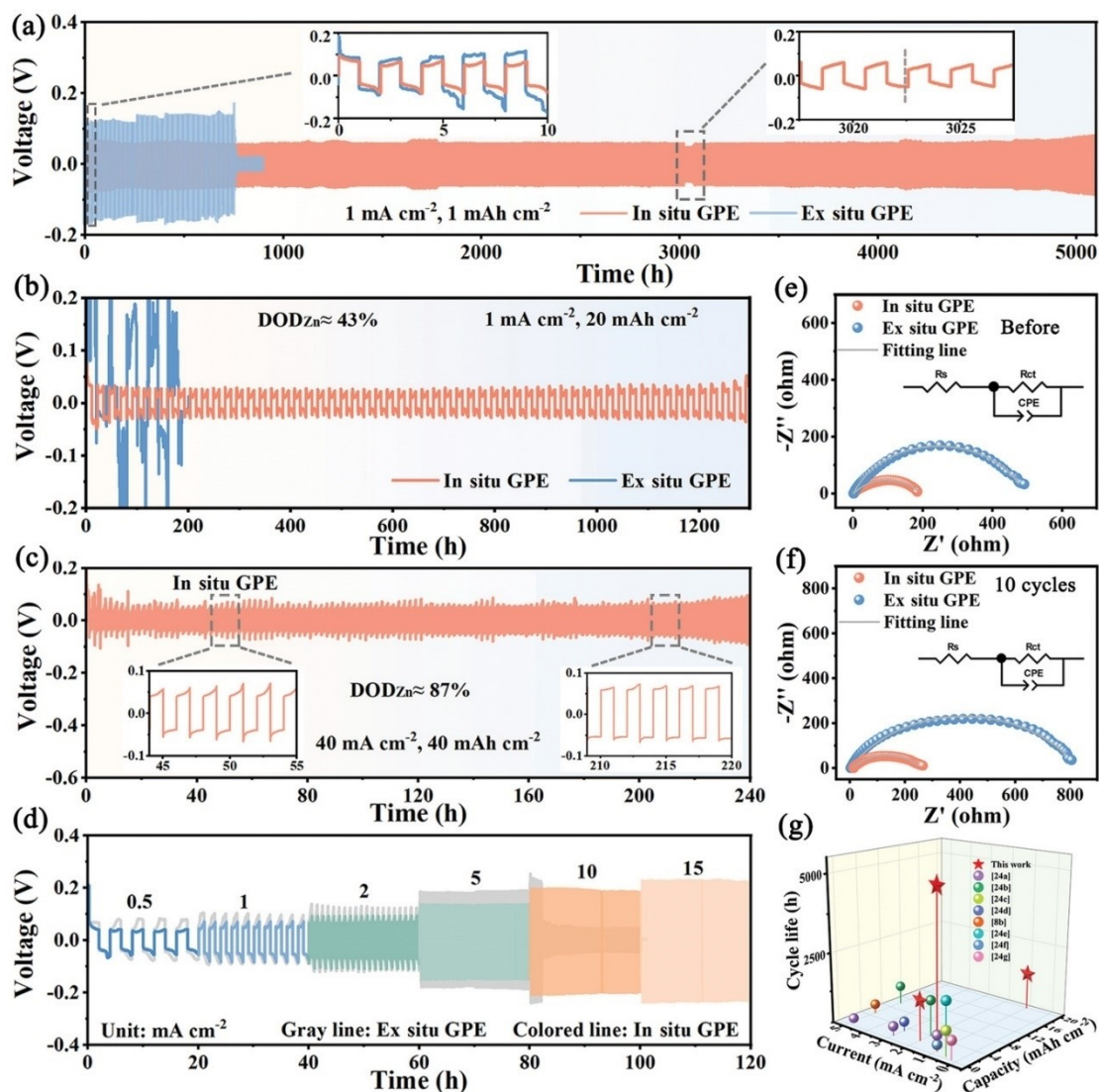


Figure 3. Electrochemical performance of the in-situ and ex-situ symmetric cells. (a–c) Long-term galvanostatic cycling performance at 1 mA cm⁻² and 1 mAh cm⁻² (d) Rate performance at different current densities with an areal capacity of 1 mAh cm⁻². (e, f) EIS curves before the cycle. (g) Comparison of cycling lifespan of symmetrical cells with reported GPEs.^[28] Reproduced from ref. [28] Copyright (2022), with the permission from Wiley-VCH.

trolyte composition optimization to improve their commercial viability.

It is important to note that these strategies are not mutually exclusive, and combinations of these approaches may be necessary to achieve the best performance for ZIBs. However, despite significant progress in mitigating side reactions in ZIBs, there is still a need for continued research in this area to improve the commercial viability of ZIBs. One area of research that requires attention is the understanding of the mechanisms behind side reactions in ZIBs. The identification of the specific chemical reactions that occur during the electrochemical processes can help researchers design better electrolytes and electrode materials to mitigate these reactions. Additionally, improving the understanding of the effect of different operating conditions, such as temperature, current density, and cycling rates, on the occurrence of side reactions is crucial for optimizing the performance of ZIBs.

Another area of research that requires attention is the development of cost-effective and scalable manufacturing processes for ZIBs with available electrolyte. Despite the low cost and abundant availability of zinc, the production of ZIBs on a large scale is still challenging. Developing cost-effective and scalable manufacturing processes electrolyte raw materials for ZIBs, such as zinc triflate, can help reduce their production costs and improve their commercial viability. Continued research in this area is crucial to improve the performance and commercial viability of ZIBs. By developing effective strategies for mitigating side reactions, researchers can improve the efficiency and lifespan of ZIBs, making them more competitive with lithium-ion batteries in a range of applications, including grid energy storage. Additionally, continued research can help identify new materials and electrolytes that are less prone to side reactions, further improving the performance and viability of ZIBs.

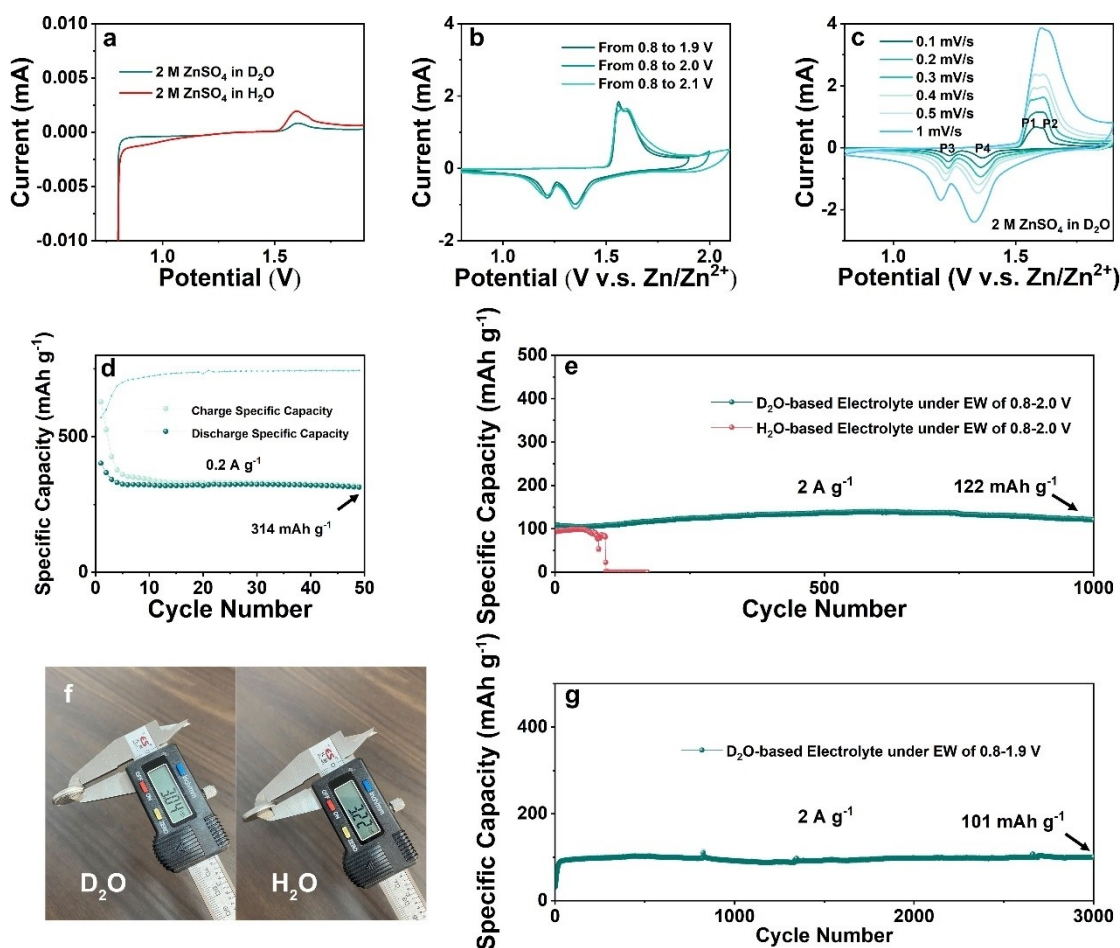


Figure 4. (a) LSV test of D₂O- and H₂O-based electrolytes. (b) CV test of D₂O-based electrolyte. (c) CV curves of D₂O-based electrolyte in the EW of 0.8–1.9 V with different scan rates. (d) Cycling performance of D₂O-based electrolytes in the EW of 0.8–2.0 V. (e) Long-term and high-rate cycling performance of D₂O-based electrolytes in the EW of 0.8–2.0 V. (f) The thickness comparison of full cells with D₂O- and H₂O-based electrolytes after cycles in the EW of 0.8–2.0 V to indicate the difference in gas generation. (g) Long-term cycling performance of the full cells with D₂O-based electrolyte in the EW of 0.8–1.9 V at a current density of 2 A g⁻¹.^[12a] Reproduced from ref. [12a] Copyright (2023), with the permission from Wiley-VCH.

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Conflict of Interests

The authors declare no conflict of interest.

Data Availability Statement

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

Keywords: zinc-ion battery · electrolyte · additives · side reaction · energy storage

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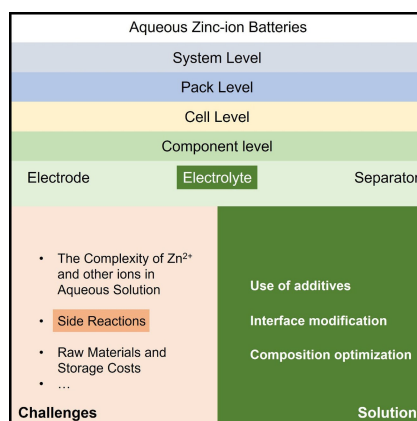
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REVIEW

Key areas of electrolytes in zinc-ion batteries (ZIBs) requiring attention include understanding the mechanisms of side reactions and developing cost-effective, scalable manufacturing processes with readily available electrolytes materials. By effectively mitigating side reactions, researchers can enhance ZIBs efficiency and lifespan, enabling them to compete with lithium-ion batteries (LIBs), particularly in grid energy storage applications.



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Recent Advances of Aqueous Electrolytes for Zinc-Ion Batteries to Mitigate Side Reactions: A Review