



Progress and Challenges in Aqueous Zinc-Ion Batteries for Optimized Full-Cell Performance

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ABSTRACT - Zinc-ion batteries (ZIBs) have recently attracted significant attention in the field of energy storage and conversion owing to their high theoretical specific capacity, low redox potential, intrinsic safety, and cost effectiveness. Despite substantial progress in performance optimization, several critical challenges still hinder their practical application, including zinc dendrite formation, cathode material instability, and relatively low operating voltage. Biomass-based materials, characterized by excellent environmental sustainability, rich surface functional groups, and tunable micro-/nanostructures, have emerged as promising candidates for the development of high-performance ZIBs. This review first introduces the chemical structures, physical properties, and synthesis strategies of biomass-derived materials. Subsequently, the configuration and working principles of zinc-ion batteries are discussed. The crucial roles of biomass-derived functional materials in zinc anodes, cathodes, electrolytes, and separators, along with their underlying electrochemical mechanisms, are systematically elucidated. Finally, the existing challenges associated with biomass-based functional materials in rechargeable ZIBs are summarized, and potential strategies and future research directions to promote their further development are proposed.

I. INTRODUCTION

With the depletion of non-renewable energy sources and the increasing prominence of environmental pollution, the development of renewable energy sources has gradually attracted widespread attention. Rechargeable Zn-ion batteries are candidates for the new generation of energy storage devices due to their high energy density, mild electrolyte, low cost and high safety performance.[1], [2], [3] To address this issue, it is essential to develop various renewable energy technologies as viable alternatives to traditional energy sources. These technologies must be supported by dependable electrochemical energy storage systems to ensure that energy usage is both environmentally friendly and sustainable. In light of this pressing requirement, Aqueous Zinc-Ion Batteries (AZIBs) present a promising solution, thanks to their superior safety features, cost-effectiveness, low flammability, high compatibility with water, and impressive theoretical specific capacity of 820 mAh/g[4], [5], [6]. Additionally, the use of mild aqueous electrolytes in Zinc-Ion Batteries (ZIBs) offers several advantages over batteries that utilize organic electrolytes, including lower costs, enhanced safety,

remarkable ionic conductivity, and a reduced environmental impact[6]. Furthermore, ZIBs represent a viable pathway for effectively utilizing renewable energy, in line with the global movement towards sustainability and minimizing carbon emission

While ZIBs offer numerous advantages, selecting an effective cathode material poses significant challenges. Currently, only a limited selection of materials, including manganese oxides, Prussian blue analogs and vanadium oxides are being explored as potential cathodes for ZIBs[6], [7], [8]. Despite the above many advantages, the actual energy density and insufficient cycle life of aqueous zinc-ion batteries are still restricting their development at this stage. Due to the narrow thermodynamic stability window of water, the voltage of zinc-ion batteries is limited, and their charging and discharging processes are always coupled with the occurrence of side reactions such as hydrogen and oxygen precipitation[9], [10], [11]. Meanwhile, it is highly influenced by the nature of electrolyte such as pH and additives, which leads to the complexity of the actual electrochemical behavior inside the battery and the unclear energy storage mechanism. Consequently, the current development of zinc-ion batteries is confronted with the following difficulties:

(1) Zinc anodes store energy through their own dissolution and deposition. They are similarly susceptible to dendrite formation during deposition, which can lead to short circuits [9], [10], [11].

(2) The majority of cathode materials exhibit a relatively low voltage plateau and are structurally unstable, prone to electrode degradation [9], [10], [11].

(3) The existing zinc storage anode materials have limited capacity, and the large polarization of divalent zinc ions slows down the kinetic processes of embedding and detachment, while destroying the structural stability of the anode material, which results in poor multiplicative performance of the battery [9], [10], [11].

(4) The electrolyte of zinc-ion batteries can be either acidic or alkaline. The occurrence of by product reactions such as hydroxide precipitation will lead to significant changes in the local pH value, in comparison with lithium-ion batteries. This places higher requirements on the acid and alkali corrosion resistance of the diaphragm.

(5) In practical outdoor use, the solidification of aqueous electrolytes at low temperatures and volatilization at high The reaction mechanism of aqueous zinc-ion batteries is controversial and has many issues compared to the reaction mechanisms of other ion batteries for energy storage. In particular, the reaction mechanism involving the energy storage process has been the focus of discussion and controversy. According to the available research papers, several reaction mechanisms have been proposed for aqueous zinc-ion batteries, including the traditional Zn^{2+} insertion/ deinsertion, dual-ion co-insertion mechanism, the chemical conversion reaction theory, and the coordination reaction of Zn^{2+} with structural organic framework cathode materials and other mechanisms [4], [5], [6]. The general categorization of the energy storage mechanisms of zinc-ion batteries and their specific reaction mechanisms and corresponding representative electrode materials will be described in detail in the following sections.

A. Zn^{2+} detachment/insertion mechanism

The Zn^{2+} detachment/insertion mechanism proposed earlier is the most common energy storage mechanism, similar to that of lithium-ion and sodium-ion batteries. During working discharge, the anode loses electrons and Zn^{2+} is embedded in the cathode active material after entering the electrolyte as a carrier, and the cathode receives electrons as the oxidation state decreases; when the battery is charged, the anode material is dissolved in the electrolyte in the form of Zn^{2+} , and electrons are obtained by the external circuit to

temperatures will accelerate the degradation of device performance [20].

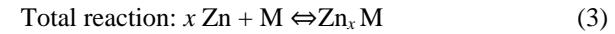
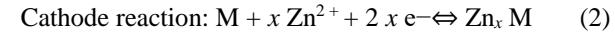
Therefore, in this review, we will start from the energy storage mechanism of zinc-ion batteries, elaborate the comparison, summarize, and analyze the energy storage mechanism of several kinds of zinc-ion batteries in detail, and then list and classify the current development status of zinc-ion batteries' anode and cathode materials, and encountered problems and the optimal solution methods. Besides, we will introduce the research status of electrolytes, and summarize the improvement methods of common types of electrolytes, aiming to make it easier for everyone to search for them. Eventually, the above aspects are summarized, and the development direction is expected. This review of zinc-ion batteries will play a role in the development of zinc-ion batteries.

II. ENERGY STORAGE MECHANISM OF ZINC-ION BATTERIES

be reduced to metallic Zn and deposited onto the anode [24], [25].

This process can be summarized in a general equation as follows:

Anode reaction:



where in M is the cathode material. Materials with tunneling, layered or open-framework structures with sufficient space to accommodate Zn^{2+} are considered ideal cathode materials for ZIBs. For example, the multistructured α - MnO_2 material was proposed by Feiyu Kang's team at Tsinghua University to reversibly insert Zn^{2+} will be inserted into the α - MnO_2 cathode/release from the α - MnO_2 cathode into the electrolyte during the discharge/charge of aqueous rechargeable zinc-ion [26]. This principle is quite different from the two-step energy storage mechanism of conventional alkaline zinc-manganese batteries. Subsequently, professor Yanbing He's team also demonstrated that the Zn^{2+} detachment/insertion mechanism occurs in β , γ , λ , δ and other types of MnO_2 . At the same time, dissolution/deposition behaviors occur on the zinc metal anode side. In alkaline conditions, the surface of metallic zinc anode is prone to passivation to generate zinc oxide passivation, and prone to generate dendrites to make

the zinc electrode utilization reduced, which is not conducive to the cyclic energy storage of zinc-ion batteries .

Challenges of the Zn anode

Generally, four factors (Table 1) influence the reversibility of Zn anodes: dendritic growth, hydrogen evolution, passivation ,and shape change.

B. Dendrite growth

Metal dendrite formation is an unavoidable problem in the battery industry. Zn dendrites are caused by the nonuniform distribution of electric fields and Zn^{2+} on the electrode surface. During charging, randomly distributed Zn^{2+} are reduced, migrated, and eventually deposited on the Zn electrode surface[. Specifically, Zn^{2+} is preferentially deposited at locations with increased Zn concentrations, forming initial tiny bumps. Subsequently, Zn deposits spontaneously at these tiny bumps due to increased surface energy, gradually evolving into Zn dendrites [27] [28]. The dendrites often display a needle-like shape, and their tips act as the charge center to induce the “tip effect” in subsequent reactions. The “tip effect” could aggravate the nonuniform distribution of the surface electric field and further cause dendrite growth. As these dendrites accumulate and grow, they lead to internal short circuits and invalidation.

Challenges for Zn-based batteries

ZN ANODE ISSUES	CAUSES	RESULTS
Dendrite growth	Uneven deposition of Zn^{2+} during charging	Dendritic growth and short circuit
Hydrogen evolution	The Zn/ZnO standard reduction potential is more negative than that of HER, leading to hydrogen evolution	Corrosion of Zn anode
Passivation	Limited mass transfer of soluble species (OH^- and $Zn(OH)_4^{2-}$) to/from the dissolving Zn anode	Decreased Zn anode utilization
Shape change	Redistribution of Zn^{2+} over the Zn anode surface during cycling	Decreased effective surface area

C. Electrolyte formulation.

The electrolyte is a crucial component in enabling high performance AZIBs. Not only does its pH indicate the reaction mechanisms at the surface, but the type of salt also has a significant impact on the electrochemical and morphological characteristics of the Zn anode. In this respect, Zhang et al. conducted a systematic investigation of

the effect of different salts and their electrochemical profiles. A survey of four mildly acidic solutions of $ZnCl_2$, $Zn(NO_3)_2$, $ZnSO_4$, and $Zn(CF_3SO_3)_2$ revealed that while nitrate- and chloride-based electrolytes were unfit for reversible deposition/dissolution, sulfate- and triflate-based solutions offered favorable environments[29].

For this process. In particular, the Zn-triflate electrolyte exhibited high stability, supposedly due to a reduced solvation effect because of the presence of bulky anions. In a high concentration of 3 M Zn ($CF_3SO_3)_2$, ex situ SEM images after galvanostatic cycling indicate a smooth morphology of Zn deposits. Ever since, the Zn ($CF_3SO_3)_2$ salt has often been employed in many studies focusing on exploring cathodes for AZIBs [30].

In the context of anode optimization, however, the Zn triflate salt alone is not enough to meet the ever-increasing expectations of the battery community in terms of performance and cycle life. Moreover, its high cost compared to $ZnSO_4$ could negate its relative electrochemical stability 32 Various alternative strategies have been explored in this regard, among which electrolyte manipulation is a popular research direction. With the common purpose of increasing the coulombic efficiency and suppressing dendrite formation, numerous formulations including triethyl phosphate (TEP),³³ polyacrylamide (PAM),³⁴ polyethyleneamine,³⁵ bio-ionic liquid,Ni triflate, SDBS, and $Zn(ClO_4)_2$ and deep eutectic solvents like acetamide[32][33].

III. RESULTS AND DISCUSSION

Recent investigations have demonstrated that biochar-assisted surface modification is an effective approach to enhance the electrochemical performance of MnO_2 cathodes for aqueous zinc-ion batteries (ZIBs). In particular, porous biochar derived from pistachio shells was utilized to form a uniform, graphitized carbon coating on MnO_2 , leading to substantial improvements in capacity, rate capability, and cycling stability. The biochar coated MnO_2 (C MnO_2) cathode delivered a high reversible specific capacity of **292.9 mA h g⁻¹ at a current density of 100 mA g⁻¹ after 50 cycles**, which is approximately three times higher than that of the pristine MnO_2 cathode ($\sim 92.5 \text{ mA h g}^{-1}$)[33].

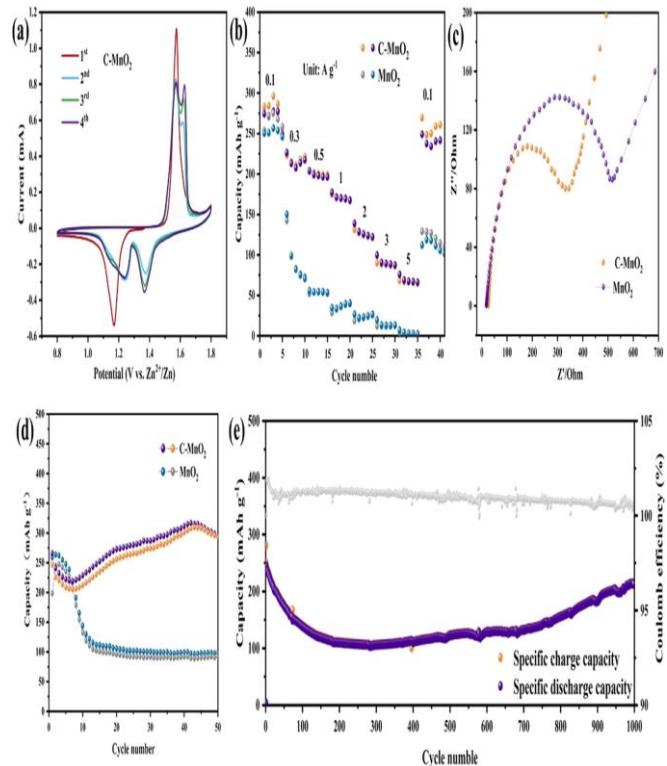
The C MnO_2 cathode also exhibited excellent rate performance over a wide range of current densities. As the current density increased from 0.1 to 5 A g⁻¹, the discharge capacity remained significantly higher than that of uncoated MnO_2 , and a capacity recovery of approximately **91%** was

achieved when the current density was returned to 0.1 A g^{-1} . Electrochemical impedance spectroscopy analysis revealed a reduced charge-transfer resistance for the biochar-coated cathode, which can be attributed to the presence of a conductive graphitized carbon network facilitating rapid electron transport.[33]

Long-term cycling evaluation further confirmed the structural and electrochemical stability imparted by the biochar coating. At a high current density of 3 A g^{-1} , the C MnO₂ cathode retained a discharge capacity of **212.8 mA h g⁻¹ after 1000 cycles**, indicating outstanding cycling durability[33]. Cyclic voltammetry analysis showed well-overlapped redox peaks after the initial activation cycles, suggesting highly reversible Zn²⁺ insertion/extraction behavior.

Kinetic analysis based on scan-rate-dependent cyclic voltammetry revealed that the charge storage mechanism of the C MnO₂ cathode is governed by a synergistic contribution of diffusion-controlled and pseudocapacitive processes. The pseudocapacitive contribution increased from approximately **45% to 67%** with increasing scan rate, highlighting the role of the porous biochar framework in enhancing ion transport kinetics and surface redox activity. Moreover, ex situ structural characterizations confirmed that the biochar coating effectively suppresses Mn dissolution and stabilizes reversible phase transformations during repeated charge discharge cycles.

Overall, these results indicate that biomass-derived biochar coatings significantly improve the electrochemical performance of MnO₂ based cathodes by enhancing electrical conductivity, accelerating reaction kinetics, and mitigating structural degradation. Such sustainable carbon modification strategies offer promising pathways for the development of high-performance and environmentally benign aqueous zinc-ion batteries.



(a) CV curves of C-MnO₂ cathode at 0.1 mV s^{-1} . (b) Rate performances of C-MnO₂ cathode and MnO₂ cathode. (c) EIS spectra of C-MnO₂ cathode and MnO₂ cathode. (d) Cycling performances of C-MnO₂ cathode and MnO₂ cathode at 0.1 A g^{-1} . (e) Long cycling performances of C-MnO₂ cathode at 3 A g^{-1} .[33]

IV. CONCLUSION

Aqueous zinc-ion batteries have attracted considerable attention as promising energy storage systems due to their high safety, low cost, and environmental compatibility. Nevertheless, their practical application is limited by challenges such as zinc dendrite formation, cathode instability, sluggish reaction kinetics, and restricted operating voltage. This review highlights recent advances in biomass-derived functional materials for zinc-ion batteries, with particular emphasis on biochar-based modifications. Biomass-derived biochar, owing to its porous structure, rich surface chemistry, and tunable conductivity, has proven to be an effective modifier for MnO₂-based cathodes. The incorporation of biochar coatings significantly enhances electrical conductivity, accelerates Zn²⁺/H⁺ transport,

suppresses manganese dissolution, and stabilizes reversible phase transformations during cycling. Consequently, biochar-assisted MnO_2 cathodes exhibit markedly improved specific capacity, rate capability, and long-term cycling stability compared to pristine MnO_2 counterparts. Electrochemical analyses further reveal that the enhanced performance originates from the synergistic contribution of diffusion-controlled and pseudocapacitive charge storage mechanisms enabled by the porous carbon framework. Despite notable progress, challenges related to large-scale fabrication, structural uniformity, and electrode–electrolyte compatibility remain. Future research should focus on rational biomass selection, precise structural regulation, and integrated electrode–electrolyte design. Overall, biomass-

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derived materials offer a sustainable and effective pathway toward the development of high-performance aqueous zinc-ion batteries for large-scale energy storage applications.

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