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ADVANCED SEPARATORS FOR AQUEOUS ZINC-ION BATTERIES

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Abstract: Aqueous zinc-ion batteries (AZIBs) are gaining traction as sustainable energy storage systems due to their safety, cost-effectiveness, and eco-friendliness. However, their practical application is hindered by critical issues such as zinc dendrite growth, hydrogen evolution reaction (HER), and cathode dissolution. The separator, a core component of AZIBs, has been increasingly recognized for its pivotal role in addressing these challenges by regulating ion transport, stabilizing interfaces, and suppressing parasitic reactions. This review systematically summarizes recent advancements in separator design and engineering strategies, including surface modifications and interface optimization. Future directions emphasize multifunctional material innovation, scalable fabrication techniques, and advanced characterization to unravel dynamic interfacial mechanisms. This work provides a comprehensive roadmap for advancing separator technology to unlock the full potential of AZIBs for grid-scale energy storage.
Keywords: Aqueous zinc-ion batteries; Separator; Dendrite-free anode

1 INTRODUCTION

Aqueous zinc-ion batteries (AZIBs) have emerged as promising candidates for large-scale energy storage due to their inherent safety, low cost, high energy density, and environmental friendliness[1, 2]. However, challenges such as zinc dendrite growth, hydrogen evolution reaction (HER), and anode corrosion severely degrade their Coulombic efficiency (CE) and cycling stability shown in Fig. 1[3]. The separator, a critical component in AZIBs, plays a pivotal role in regulating ion transport, stabilizing interfaces, and suppressing side reactions[4,5]. Recent advancements in separator design and functionalization have significantly improved AZIB performance, making this a key area of research.

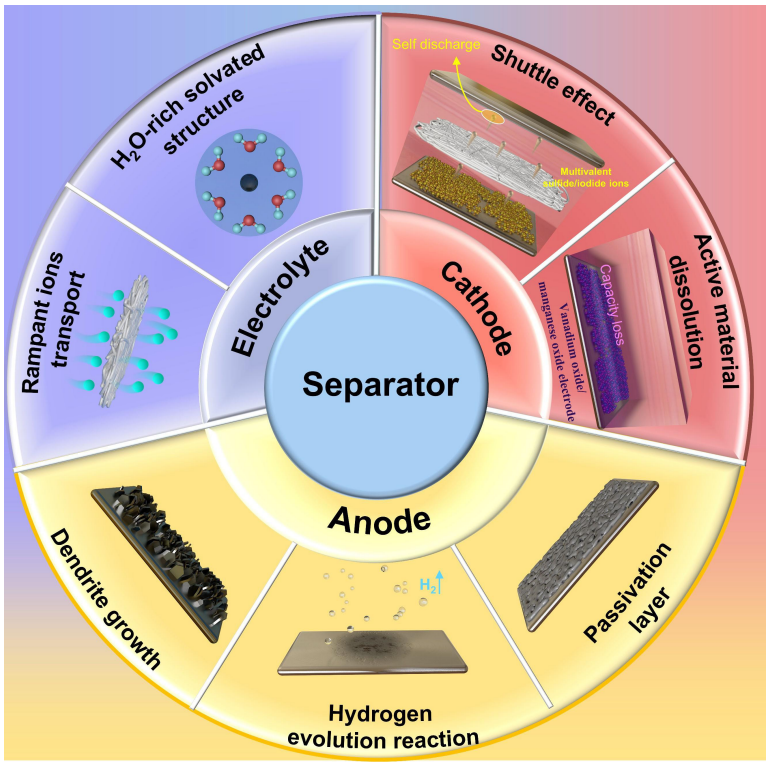

Figure 1 Comprehensive Examination of Challenges Associated with AZIBs, Encompassing Concerns Related to the Electrolyte, Cathode, and Anode[3]

2 OPTIMIZATION STRATEGIES FOR SEPARATORS

The performance of the separator dictates the battery's interfacial configuration and internal resistance, thereby influencing overall battery functionality, encompassing capacity, cycle stability, and safety as shown in Fig. 2.[6] For

zinc-based energy devices, glass fiber separators are widely used due to their low electrical conductivity, appropriate porosity, high ionic conductivity, and good wettability with aqueous electrolytes.[7] However, their internally disordered pore size distribution and rough surface often lead to irregular electroplating of zinc, resulting in the formation of dendrites.

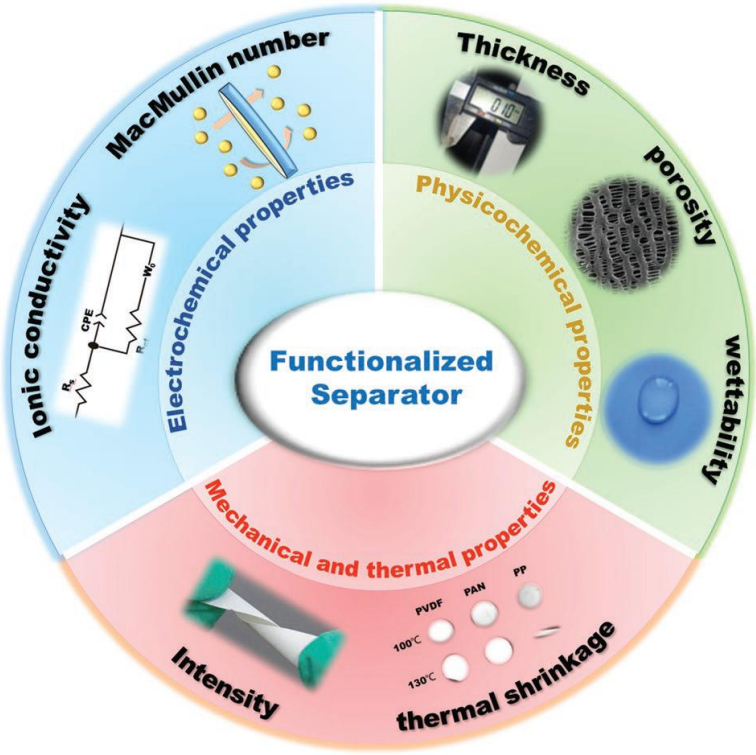

Figure 2 Illustrative Representation Outlining the Critical Characteristic Demands for Functionalized Separator[6]

2.1 Surface Modification

Recent research has found that functionalizing the surface of the separator can effectively regulate the transport of Zn^{2+}. Traditional glass fiber (GF) or cellulose separators are modified to improve wettability and ion transport. For instance, spraying a conductive KB layer on GF enhances electrolyte affinity and accelerates Mn^{2+} electrooxidation to form electrochemically active birnessite, balancing capacity fading[8].

2.2 Functional Interlayers

Introducing interlayers between the separator and electrodes can redistribute ion flux[9]. Zr-based MOFs (e.g., UiO-66) embedded in GF homogenize pore structures, confine zinc ions, and suppress dendrites via size-selective ion transport[10].

2.3 Interface Engineering

Separators optimize electrode/electrolyte interfaces (EEI) by regulating solvation structures. For example, hydrophilic functional groups on separators adjust Zn^{2+} coordination, reducing free water activity and HER[3, 6].

3 MECHANISM ANALYSIS OF SEPARATOR ON BATTERIES

As a core component for ion transport and interface regulation in aqueous zinc-ion batteries (AZIBs), the modification strategy of the separator significantly extends the battery cycle life by optimizing zinc deposition behavior, suppressing side reactions, and enhancing interface stability[11]. The following systematically elaborates on the key mechanisms of membrane modification, combined with the latest research findings:

3.1 Regulation of Ion Transport and Electric Field Homogenization

The separator can effectively regulate the diffusion path and concentration distribution of zinc ions by optimizing the pore structure and surface functional groups, thereby inhibiting dendrite growth caused by excessive local current density[12, 13]. For example: Gradient pore design: MOF-based composite separators construct gradient pore structures through hot pressing, using the high specific surface area (190.8 m^2/g) and strong adsorption of Zn^{2+} by nitrogen functional groups of MOF to dynamically regulate ion flux. Experiments show that this separator can achieve a cycle life of 3000 h for Zn||Zn symmetric cells and induce uniform deposition of zinc along the (002) crystal plane[14].

Zwitterionic modification: PAN@SBMA separators regulate zinc ion concentration polarization with sulfonic acid groups (SO_3-), homogenizing the electric field distribution. The ion transfer number ($t(Zn^{2+})$) is significantly improved, enabling stable cycling of Zn||Zn symmetric cells for 1700 hours at 1 mA/cm², far exceeding traditional glass fiber separators[15].

3.2 Crystal Facet Regulation and Heterogeneous Epitaxy

Functionalization of the separator surface can induce preferential orientation of zinc deposition crystal facets, suppressing the formation of disordered dendrites[16]:
MOFs induce crystal facet regulation: UiO-66-GF separators use the porous confinement effect of Zr-based MOFs to induce preferential deposition of zinc along the (002) crystal plane. DFT calculations show that the (002) crystal plane has a lower adsorption energy for H (-1.731 eV), effectively suppressing the hydrogen evolution reaction (HER) and corrosion[10].

3.3 Interface Chemistry Optimization and Side Reaction Suppression

The separator reduces contact between active water molecules and the zinc anode through interface chemical modification, suppressing side reactions:
Hydrophobic/hydrophilic synergistic design: P/FS-Z separators combine a hydrophobic PTFE matrix with hydrophilic SiO_2 nanofillers, reducing the activity of water in the electrolyte. FTIR characterization shows a redshift of the O-H vibration peak (1634.4 cm^{-1}→1630.1 cm^{-1}), confirming that the activity of water molecules is suppressed, thereby reducing HER and corrosion reactions.
Dynamic concentration regulation: MOF-modified separators dynamically adsorb zinc ions with nitrogen functional groups, promoting two-dimensional grain growth at low concentrations and inducing uniform nucleation at high concentrations, achieving phased regulation of zinc deposition and significantly suppressing the "tip effect".

3.4 Mechanical Reinforcement and Multi-scale Synergy

Enhancing the mechanical properties of the separator can prevent dendrite penetration, while multi-scale synergistic design enhances interface stability:
Nanofiber reinforcement: Bacterial cellulose separators, with a high porosity rate (>90%) and an interwoven nanofiber structure[17,18].

4 FUTURE PERSPECTIVES

1. Material Innovation: Explore multifunctional frameworks (e.g., covalent organic frameworks) for synergistic ion regulation and HER suppression.
2. Scalable Fabrication: Develop low-cost, high-throughput methods for industrial adoption[6].
3. Multidimensional Design: Integrate separators with smart responsiveness (e.g., self-healing or pH regulation) [19].
4. Mechanistic Studies: Advanced in situ characterization to unravel dynamic interfacial processes.

5 CONCLUSION

Advanced separator engineering has become a cornerstone for high-performance AZIBs, addressing dendrites, HER, and cathode degradation. By leveraging surface modifications, functional interlayers, and novel materials, researchers have achieved remarkable improvements in cycling stability and energy density. Future efforts should focus on scalable solutions and holistic system design to accelerate commercialization.

COMPETING INTERESTS

The authors have no relevant financial or non-financial interests to disclose.

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