Aqueous Zinc-Ion Batteries: A Survey

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Abstract

Aqueous zinc-ion batteries (AZIBs) represent a promising avenue for sustainable energy storage, offering cost-effectiveness and safety. This survey paper systematically explores the challenges and advancements associated with AZIBs, focusing on the zinc metal anode and the critical issue of dendrite formation. Dendrite suppression is essential to prevent short circuits and enhance battery longevity, with strategies such as 'soggy-sand electrolytes' and interfacial engineering showing promise. Electrolyte innovations, particularly water-in-salt systems, are pivotal in improving ionic conductivity and stability, while polymer coatings mitigate corrosion and promote uniform zinc deposition. The paper also delves into ion transport mechanisms, emphasizing the role of inorganic solid electrolytes and advanced modeling techniques in optimizing ionic conductivity. Recent advancements in anode materials and future research directions highlight the potential for significant improvements in AZIB performance. Continued research is crucial to overcoming existing challenges, ensuring the development of high-performance, durable, and cost-effective AZIBs to meet the demands for sustainable energy storage solutions.

1 Introduction

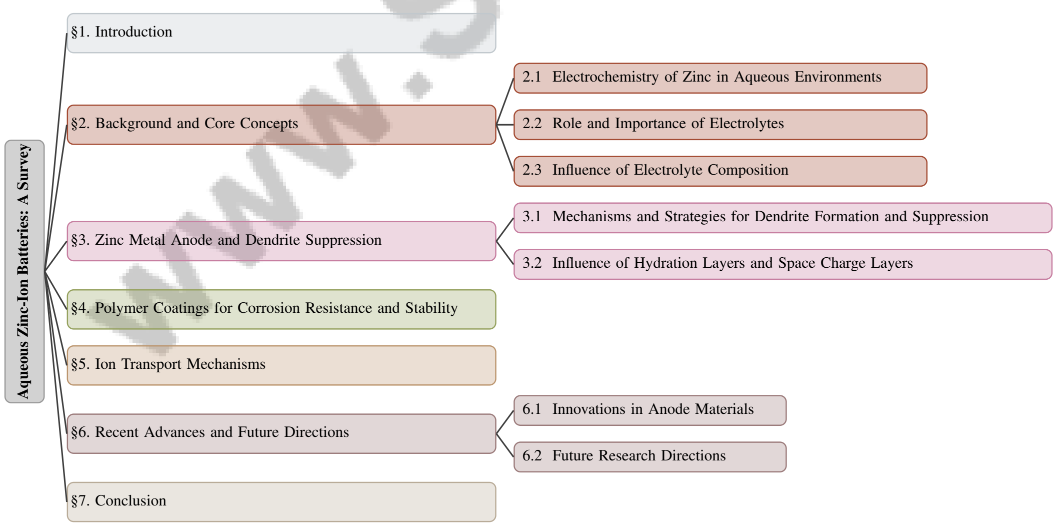

Figure 1: chapter structure

1.1 Significance of Aqueous Zinc-Ion Batteries

Aqueous zinc-ion batteries (AZIBs) represent a promising advancement in energy storage technology, offering low-cost, environmentally friendly, and safe alternatives to traditional batteries [1]. Their significance lies in addressing critical energy storage challenges, particularly concerning the zinc metal anode, which provides intrinsic safety and cost-effectiveness essential for large-scale stationary applications [2].

However, AZIBs encounter obstacles such as limited electrochemical stability windows, side reactions, and zinc dendrite growth, which can compromise performance and safety [3]. Issues like corrosion and random dendrite formation further diminish battery efficiency and longevity [4]. Tackling these challenges is crucial for enhancing AZIB performance and ensuring their competitiveness in the energy storage market [5].

Research emphasizes the critical role of zinc metal anodes in AZIBs, particularly in mildly acidic environments, where understanding their electrochemical behavior is essential [6]. Effective management of dendrite formation and corrosion through innovative strategies could facilitate the widespread adoption of AZIBs [7]. Thus, the development of AZIBs not only advances energy storage technologies but also contributes to a sustainable and secure energy future [8].

1.2 Structure of the Survey

This survey is systematically structured to provide a comprehensive exploration of AZIBs and the multifaceted challenges they pose, particularly focusing on zinc dendrite growth and mitigation strategies [8]. The introduction highlights the significance of AZIBs as a viable energy storage solution and outlines the challenges related to zinc metal anodes. Following this, the background and core concepts section discusses the fundamental principles governing zinc electrochemistry in aqueous environments and the critical role of electrolytes in battery performance.

The subsequent section addresses the zinc metal anode and the pressing issue of dendrite suppression, examining the mechanisms of dendrite formation and innovative suppression strategies. An in-depth analysis of polymer coatings is presented, emphasizing their role in enhancing corrosion resistance and electrochemical stability of zinc anodes. Additionally, the survey explores the integration of polymer coatings with electrolyte innovations to further improve battery performance.

The discussion advances to ion transport mechanisms, analyzing their dynamics within AZIBs and their impact on overall efficiency. The section on recent advances and future directions highlights innovations in anode materials and polymer coatings while identifying key areas for future research. The conclusion synthesizes the key findings, underscoring the necessity of addressing dendrite formation and enhancing ion transport to propel the development of efficient AZIBs.The following sections are organized as shown in Figure 1.

2 Background and Core Concepts

2.1 Electrochemistry of Zinc in Aqueous Environments

The electrochemical performance of zinc in aqueous environments is significantly impacted by the constraints of aqueous electrolytes, particularly their limited electrochemical stability window, which induces undesirable side reactions and impedes efficient ion transport [3]. A critical issue is the low plating and stripping efficiency of zinc, further complicated by dendrite formation, which can lead to short circuits [1]. Additionally, the spontaneous zinc-water reaction produces hydrogen gas, diminishing effective battery capacity [1].

Zinc's thermodynamic instability during electroplating and stripping exacerbates dendrite growth and parasitic reactions, presenting substantial challenges for high-performance aqueous zinc-ion batteries [5]. Addressing these issues requires advanced interfacial engineering strategies, such as surface modifications and electrolyte additives, to improve zinc anode performance [7]. Understanding zinc's electrochemical stability in relation to potential cathode materials is crucial. The refined Pourbaix diagram is instrumental in determining optimal conditions for aqueous Zn/MnO2 rechargeable cells, facilitating the exploration of new cathode materials [9]. The performance of zinc-ion batteries is also hindered by the limited efficacy of existing MXenes as cathodes, due to inadequate surface terminal modifications [10].

Furthermore, the intrinsic limitations of ionic conductivity in polymer electrolytes, especially dry single-ion conducting polymers, pose additional challenges [11]. Innovative methods to enhance corrosion resistance and electrochemical stability are essential for advancing efficient aqueous zinc-ion batteries. Theoretical frameworks for ion transport dynamics can be enriched through nonequilibrium statistical mechanics and the BBGKI chain of equations, which describe interacting particle kinetics [12].

2.2 Role and Importance of Electrolytes

Electrolytes play a pivotal role in the design and operation of aqueous zinc-ion batteries (AZIBs), facilitating ion transport and stabilizing electrode-electrolyte interfaces [13]. The electrolyte composition significantly affects electrochemical performance and battery lifespan. Incorporating insulating oxide particles, as in 'soggy-sand electrolytes,' reduces water activity, enhances Zn^{2+} transport, and improves zinc deposition uniformity [3].

Exploring water-in-salt electrolytes, such as circumneutral concentrated ammonium acetate solutions, offers safety and cost benefits while maintaining strong electrochemical performance [14]. However, challenges remain with the corrosive nature of halide salts in near-neutral electrolytes, which can impair battery performance and longevity, particularly in zinc-air systems [15]. Rapid degradation in AZIBs is often due to poor zinc anode reversibility and oxide cathode dissolution, highlighting the need for more stable electrolyte compositions [2]. Enhancing MXenes' electrochemical performance can be achieved by exploring new halogenated surface terminals for improved electrolyte interactions [10].

Understanding asymmetries in ion valences and diffusivities is essential for optimizing ion transport within electrolytes, especially in charged cylindrical pores [16]. The low ionic conductivity of dry polymer electrolytes at ambient temperatures presents another challenge, necessitating innovative strategies to enhance ion jump rates for effective conductivity [11]. Addressing these challenges through electrolyte innovations is critical for advancing the performance and reliability of AZIBs.

2.3 Influence of Electrolyte Composition

Electrolyte composition is crucial in determining the performance and stability of aqueous zinc-ion batteries (AZIBs). Optimizing electrolyte mixtures is vital for enhancing ionic conductivity while minimizing experimental complexity and costs [13]. A significant challenge in conventional electrolytes is the presence of free water molecules, which can lead to side reactions and non-uniform zinc plating, adversely affecting battery performance [2].

To mitigate these issues, researchers are exploring water-in-salt electrolytes, such as circumneutral concentrated ammonium acetate solutions, which balance high ionic conductivity with wide electrochemical stability windows, ensuring safety and environmental friendliness [14]. These innovations aim to reduce the adverse effects of free water molecules and enhance zinc deposition uniformity.

Recent advancements in electrolyte design have utilized direct numerical simulations to solve Poisson-Nernst-Planck equations, effectively capturing the influence of asymmetries in ionic diffusivities and valences on ion transport [16]. This approach provides deeper insights into ion transport dynamics, enabling the development of more effective electrolyte compositions that enhance the overall performance and stability of AZIBs.

In recent years, the focus on zinc metal anodes has intensified due to their potential in next-generation batteries. However, challenges in zinc electrodeposition have emerged as significant barriers to their practical application. To address these challenges, researchers have developed various strategies and mechanisms aimed at dendrite suppression. Figure 2 illustrates the hierarchical structure of these strategies, showcasing the interplay between countermeasures, the role of interfacial engineering, and the influence of hydration and space charge layers. This comprehensive overview not only highlights the complexity of the issue but also underscores the importance of a multifaceted approach to enhance the performance and longevity of zinc anodes.

3 Zinc Metal Anode and Dendrite Suppression

3.1 Mechanisms and Strategies for Dendrite Formation and Suppression

Dendrite formation during zinc electrodeposition in aqueous zinc-ion batteries (AZIBs) significantly impedes achieving high energy density and reliability [17]. These structures can penetrate the separator, causing internal short circuits and reducing battery lifespan [17]. The instability of zinc

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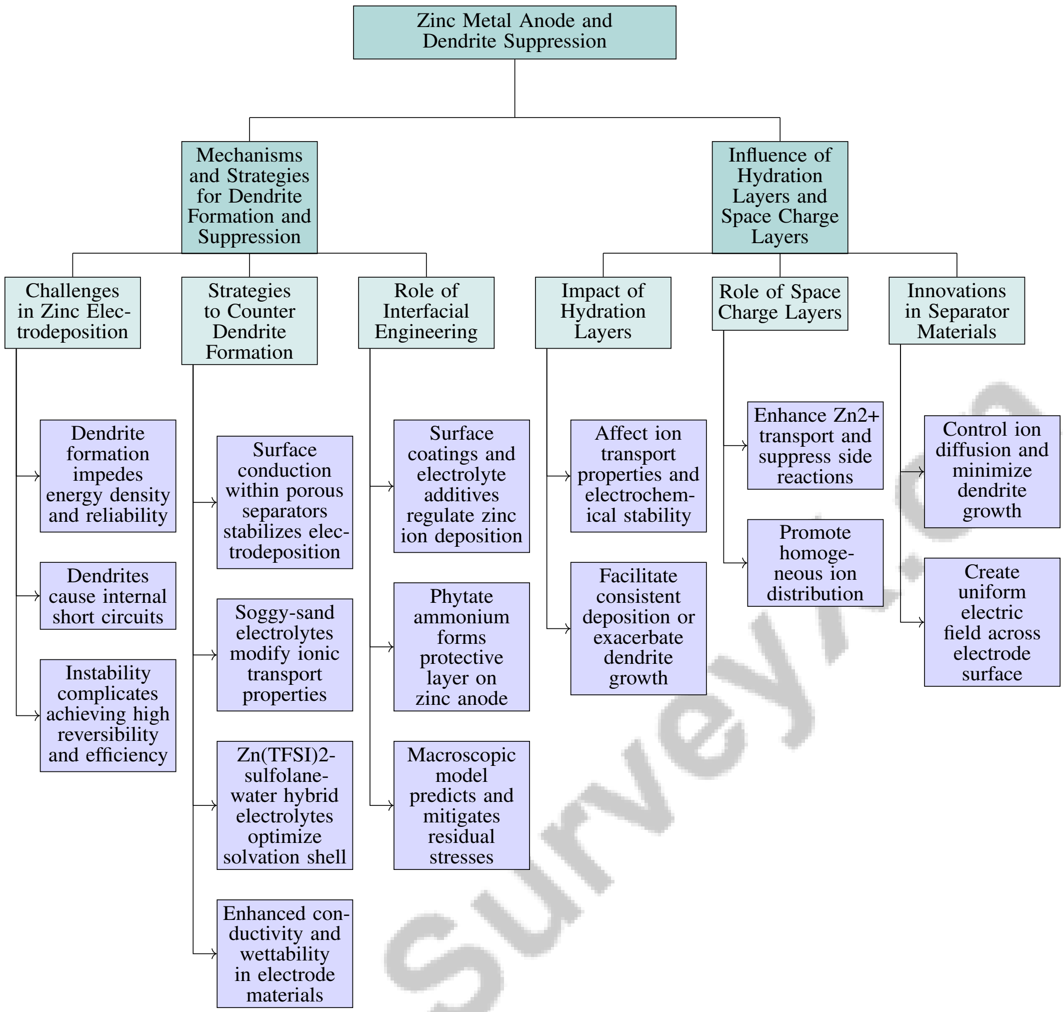


Figure 2: This figure illustrates the hierarchical structure of strategies and mechanisms for dendrite suppression in zinc metal anodes, focusing on challenges in zinc electrodeposition, counter strategies, the role of interfacial engineering, and the influence of hydration and space charge layers.

deposition is further complicated by challenges in achieving high reversibility and efficiency essential for optimal performance [6].

To counter these issues, several strategies have been developed. Surface conduction within charged porous separators stabilizes electrodeposition, suppressing dendritic growth and enhancing electrochemical performance [17]. 'Soggy-sand electrolytes' modify ionic transport properties, promoting uniform zinc deposition [3]. The use of $Zn(TFSI)_2$-sulfolane-water hybrid electrolytes optimizes the solvation shell, reducing dendrite formation and improving deposition stability [5]. Enhanced conductivity and wettability in electrode materials also contribute to uniform zinc deposition, mitigating dendrite formation [8].

Interfacial engineering, including surface coatings and electrolyte additives, plays a crucial role in regulating zinc ion deposition. Phytate ammonium (PAN) serves as a multifunctional additive, forming a protective layer on the zinc anode to enhance ion deposition uniformity and suppress dendrite growth [4]. Additionally, residual stresses from lattice misfit during deposition can negatively affect anode performance [18]. A macroscopic model combining ab initio simulations with continuum theories can predict and mitigate these stresses, enhancing anode reliability [18].

As illustrated in Figure 3, the mechanisms of dendrite formation and various strategies for their suppression in aqueous zinc-ion batteries are highlighted, showcasing key methods such as surface conduction, soggy-sand electrolytes, and interfacial engineering techniques. Innovative techniques and comprehensive engineering strategies are vital for overcoming dendrite formation in AZIBs, ensuring improved performance and longevity. The kinetic description of ion transport in systems with ionic solutions interacting with porous media, especially in nonequilibrium scenarios, enriches the theoretical framework necessary for advancing these strategies [12].

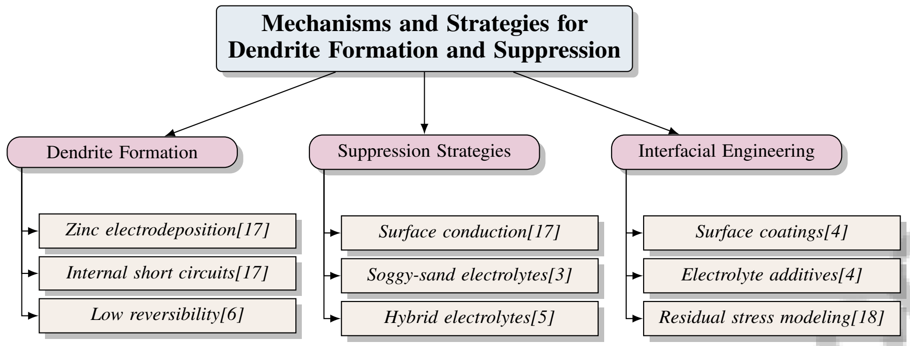


Figure 3: This figure illustrates the mechanisms of dendrite formation and various strategies for their suppression in aqueous zinc-ion batteries, highlighting key methods such as surface conduction, soggy-sand electrolytes, and interfacial engineering techniques.

3.2 Influence of Hydration Layers and Space Charge Layers

Hydration and space charge layers significantly impact dendrite formation and zinc metal anode performance in AZIBs. Hydration layers, formed by water molecule adsorption on electrode surfaces, affect ion transport properties and electrochemical stability. The structural characteristics and interactions of these layers with electrolyte components can either facilitate consistent deposition or exacerbate dendrite growth and parasitic reactions, impacting battery performance and longevity [4, 2, 6].

Space charge layers, formed around oxide particles in the electrolyte, enhance Zn^{2+} transport and suppress undesirable side reactions [3]. By altering local electric fields and concentration gradients, these layers promote a more homogeneous ion distribution, crucial for minimizing dendrite growth and improving battery performance.

Innovations in separator materials are crucial for controlling ion diffusion and minimizing dendrite growth by leveraging space charge effects [8]. Advanced separators create a uniform electric field across the electrode surface, reducing localized zinc deposition that leads to dendrite formation.

The interplay between hydration and space charge layers is essential for optimizing zinc ion deposition on the anode surface, crucial for enhancing cycling stability and overall performance of rechargeable aqueous zinc-ion batteries (RAZIBs). Engineering these layers can achieve a balance that promotes uniform zinc plating, enhances ionic conductivity, and improves AZIB efficiency and longevity. Understanding and manipulating these interfacial phenomena are key to advancing high-performance zinc-ion batteries [7, 6].

4 Polymer Coatings for Corrosion Resistance and Stability

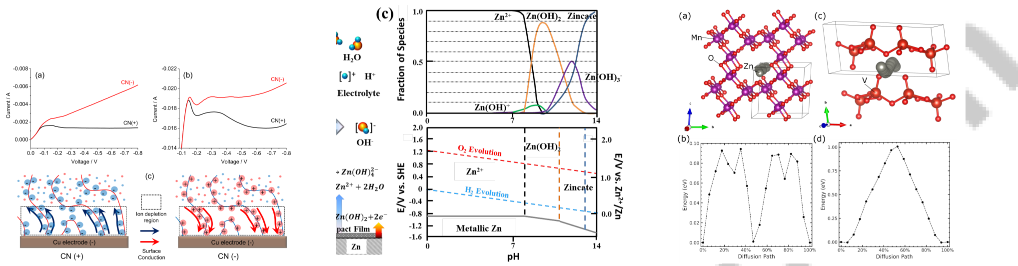
The exploration of polymer coatings is pivotal in advancing energy storage technologies, particularly in enhancing the corrosion resistance and electrochemical stability of zinc metal anodes in aqueous zinc-ion batteries (AZIBs). The following subsection examines the specific roles of polymer coatings and the innovative techniques being developed to maximize their effectiveness, providing insights into how these coatings contribute to the performance and reliability of AZIBs, especially when integrated with electrolyte innovations.

4.1 Role of Polymer Coatings and Innovative Techniques

Polymer coatings significantly enhance the corrosion resistance and electrochemical stability of zinc metal anodes in AZIBs by forming protective barriers that minimize direct contact with aqueous electrolytes, thereby reducing side reactions and ensuring uniform zinc deposition [8]. These coatings improve cycling stability and efficiency by mitigating zinc dendrite formation and corrosion.

Innovative techniques, such as the design of halide-free electrolytes using organic salts, offer promising enhancements to battery performance while avoiding the corrosive effects typical of halide-based systems [15]. These approaches not only stabilize electrolytes but also complement polymer coatings, resulting in robust and efficient battery systems.

Integrating modified electrodes, additives, and advanced separators with polymer coatings further enhances AZIB performance by improving anode interfacial properties, promoting uniform ion transport, and reducing dendrite formation [8]. Such combined strategies significantly extend the longevity and reliability of AZIBs, facilitating broader applications in energy storage solutions.



(a) Electrochemical Impedance Spectroscopy (EIS) Analysis of Copper Electrode in Presence of Cyanide Anions[17]

(b) The image depicts a graph and diagram related to zinc corrosion and electrochemical reactions.[1]

(c) Diffusion Path Energy in MnO-ZnO-VO2 System[19]

Figure 4: Examples of Role of Polymer Coatings and Innovative Techniques

Figure 4 illustrates the crucial role of polymer coatings in enhancing corrosion resistance and stability by forming protective barriers. Electrochemical Impedance Spectroscopy (EIS) provides insights into the efficacy of these coatings, as shown by the analysis of copper electrodes with cyanide anions, highlighting impedance changes. Understanding zinc corrosion through detailed graphs of species fractions as a function of pH underscores the importance of polymer coatings. Additionally, diffusion path energy studies in systems like MnO-ZnO-VO2 reveal the molecular-level stability provided by these coatings [17, 1, 19].

4.2 Integration with Electrolyte Innovations

The integration of polymer coatings with electrolyte innovations is vital for enhancing the stability and performance of AZIBs. Polymer coatings mitigate corrosion and dendrite formation on zinc anodes, while electrolyte innovations optimize ionic conductivity and electrochemical stability. Advanced electrode designs, high-concentration electrolytes, and interfacial engineering strategies collectively enhance AZIB efficiency and lifespan, addressing challenges like dendrite formation and cycling performance degradation [4, 2, 7, 8].

Recent advancements, such as water-in-salt electrolytes and insulating oxide particles, reduce water activity and enhance Zn^{2+} transport, complementing polymer coatings by stabilizing the electrochemical environment and promoting uniform zinc deposition [3]. The DiffMix model provides a novel approach to optimizing electrolyte compositions for fast-charging applications, enabling rapid identification of optimal formulations that enhance polymer-coated zinc anode performance [13].

Furthermore, integrating polymer coatings with halide-free organic salt-based electrolytes reduces zinc anode corrosion, extending battery life and performance [15]. These combined innovations offer robust energy storage solutions, paving the way for broader AZIB adoption across various applications.

5 Ion Transport Mechanisms

Understanding ion transport mechanisms is crucial for improving zinc-ion battery performance, as these processes dictate ionic movement within materials. A detailed examination of ionic conductivity

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is vital, impacting the efficiency of electrolytes like single-ion conducting polymers and inorganic solid electrolytes, which are increasingly used in advanced energy storage technologies [11, 20]. This section focuses on the role of ionic conductivity in inorganic solid electrolytes and polymers, both of which are essential for optimizing zinc-ion battery performance.

5.1 Ionic Conductivity in Inorganic Solid Electrolytes and Polymers

Ionic conductivity in materials for zinc-ion batteries is a key factor in efficient ion transport and overall battery performance. Inorganic solid electrolytes (ISEs) such as LISICON-like, NASICON-like, perovskite/anti-perovskite, and garnet electrolytes offer varying ionic conductivities, crucial for their application in energy storage devices [20]. These materials are preferred for their high ionic conductivity and stability, enhancing battery performance.

Advanced simulation techniques, including all-atom molecular dynamics and umbrella sampling, provide insights into the free energy profiles associated with ion entry into nanostructures like carbon nanotubes [21]. Such simulations are essential for understanding ion transport mechanisms at the molecular level, guiding the design of more efficient electrolytes.

The Unified Theory of Overlimiting Current (OLC) in microchannels highlights the interplay between surface charge and ion transport dynamics, paralleling the examination of ionic conductivity in battery materials [22]. This theory underscores the importance of surface interactions in modulating ion transport, offering insights for optimizing electrolyte formulations.

The coupling of charge and salt transport processes is influenced by asymmetries in ionic diffusivities and valences, as demonstrated through direct numerical simulations [16]. These asymmetries can alter transport dynamics, necessitating precise modeling for optimal electrolyte performance.

Challenges remain in understanding the complex interactions between ions and the porous structure of electrolytes, including spatial heterogeneity and accurate diffusion coefficient modeling [12]. Addressing these challenges through innovative approaches and advanced modeling techniques is essential for developing high-performance zinc-ion batteries, ensuring efficient ion transport and enhanced battery efficiency.

5.2 Modeling Ion Transport Dynamics

Modeling ion transport dynamics in zinc-ion batteries is vital for optimizing battery performance. Advanced modeling approaches capture the intricate interactions governing ion transport. One approach integrates orientational weight terms into superposition models, enhancing the representation of hydration dynamics and ion transport [21]. This modification improves model accuracy by considering the directional dependencies of ion movement, refining predictions of ion transport behavior.

Theoretical models elucidate the impact of point defects and structural arrangements on ionic conductivity and transport mechanisms [20]. These models reveal how variations in material structure influence ion transport pathways and efficiency, providing insights for material design and optimization.

A comprehensive understanding of ion transport dynamics requires integrated models that account for both molecular-level interactions—such as ion hydration and structural changes—and macroscopic phenomena, including porous environments and electrokinetic behaviors, critical for applications in energy storage, desalination, and biosensing technologies [20, 12, 21, 22, 16]. These models must incorporate the interplay between charge transport and structural features, such as porosity and surface charge, to accurately simulate ion behavior within the battery system.

Employing sophisticated modeling techniques enhances the predictive accuracy of zinc-ion battery performance across varying operating conditions, facilitating the identification of optimal materials and electrolyte formulations that significantly improve ion transport efficiency and overall battery performance. For instance, computational screening of cathode materials evaluates critical factors like the feasibility of Zn^{2} intercalation, thermodynamic stability, and electrochemical stability, while advancements in concentrated dual-cation electrolytes have shown effectiveness in mitigating performance degradation in aqueous zinc-ion batteries, promoting better cycling stability and rate performance [2, 19].

6 Recent Advances and Future Directions

6.1 Innovations in Anode Materials

Recent progress in anode materials has significantly enhanced the performance and lifespan of aqueous zinc-ion batteries (AZIBs). Techniques like shock electrodeposition have proven effective in mitigating dendritic growth, promoting uniform metal deposition in porous media, and thus improving cycle life and performance [17]. Alongside electrodeposition, computational screening has identified promising cathode materials such as MoO_3 and FeO_2, despite their stability challenges, underscoring the need for further optimization [19]. Additionally, advancements in electrolyte technology, including the development of halide-free aqueous electrolytes using thermodynamic descriptors for component screening, have enhanced the electrochemical environment, reducing corrosive effects and supporting more stable anode materials [15].

These innovations address critical issues like dendrite formation and side reactions that have historically impeded zinc metal anodes. Strategies such as interfacial engineering have resulted in more uniform zinc deposition and improved cycling stability, positioning AZIBs as a viable alternative to lithium-ion batteries due to their environmental benefits, cost-effectiveness, and high theoretical capacity [6, 1, 7, 8]. Ongoing research is vital for overcoming existing challenges and fully realizing the potential of AZIB technologies.

6.2 Future Research Directions

The future of AZIBs hinges on research aimed at overcoming current challenges and enhancing battery performance. Optimizing dendrite suppression techniques, such as shock electrodeposition, across various materials and conditions is crucial for broader applicability [17]. Efforts to reduce zinc anode weight while maximizing reversibility and energy density are essential, necessitating exploration of innovative host materials and novel electrolytes [6]. Refining 'soggy-sand electrolytes' and assessing their applicability in other battery systems, such as lithium-ion and sodium-ion batteries, present promising research avenues [3].

Enhancing models to account for residual stress during deposition, integrating thermal effects, and varying conditions is vital for predicting and mitigating stress impacts on zinc anodes [18]. Advances in sampling methods for water orientation distribution and studies on ion transport in diverse environments, such as charged nanopores, are critical for understanding hydration dynamics and ion mobility [21]. Optimizing the synthesis and structural properties of inorganic solid electrolytes (ISEs), exploring new materials, and addressing interfacial stability challenges are essential for improving ionic conductivity in AZIBs [20].

Exploring the scalability of additives like phytate ammonium and their long-term effects on battery performance, alongside optimizing electrolyte compositions, merits detailed investigation [4]. Developing closed-loop optimization strategies that adaptively retrain models based on experimental feedback and exploring alternative physical laws for ionic conductivities are promising for enhancing AZIB performance [13]. Research into hybrid systems incorporating redox-active materials and alternative materials to mitigate degradation during charging cycles is critical for boosting energy performance.

Extending models to incorporate chemical effects and examining their applicability in various configurations are crucial for advancing AZIB technology [22]. Future research should consider real-world applications, such as energy storage and environmental technologies, and refine models to include detailed interactions validated with experimental data to enhance understanding and development of AZIBs [16, 12]. These directions are pivotal for advancing AZIB technology and facilitating its broader adoption in energy storage solutions.

7 Conclusion

The exploration of aqueous zinc-ion batteries (AZIBs) underscores their potential as a sustainable and effective solution for energy storage, while also highlighting the critical challenges and advancements that shape the field. Central to these challenges is the zinc metal anode, where the formation of dendrites poses significant risks to both performance and safety. Addressing this issue through innovative suppression techniques, such as 'soggy-sand electrolytes' and advanced interfacial engineering, is crucial for enhancing the operational reliability and longevity of AZIBs.

The evolution of electrolyte compositions, notably the development of water-in-salt systems, has been pivotal in enhancing ionic conductivity and electrochemical stability. The strategic use of polymer coatings plays a complementary role by reducing corrosion and ensuring uniform zinc deposition, thereby contributing to a holistic improvement in battery performance.

Furthermore, a deeper understanding of ion transport mechanisms within AZIBs highlights the importance of optimizing ionic conductivity. This can be achieved through the use of inorganic solid electrolytes and sophisticated modeling techniques, which are essential for achieving efficient ion transport and enhancing overall battery efficiency.

Innovations in anode materials, coupled with ongoing research into electrolyte and coating technologies, are poised to drive significant improvements in AZIB performance. Continued research efforts are vital to overcoming the existing challenges, paving the way for the development of high-performance, durable, and cost-effective AZIBs capable of meeting the growing demand for sustainable energy storage solutions.

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