Proceedings of CONF-MSS 2026 Symposium: Mechanical Control and Automation
DOI: 10.54254/2755-2721/2026.BJ31522

Research Progress on the Application of Graphene Materials in Aqueous Zinc-ion Batteries

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Abstract. Aqueous zinc-ion batteries have shown great potential in large-scale energy storage fields for their merits like high safety, environmental friendliness and low cost. However, the dendrite growth and hydrogen evolution corrosion of the zinc anode, as well as the poor intrinsic conductivity and unstable structure of cathode materials, restrict its electrochemical performance and cycle life. Owing to its exceptional characteristics—including ultra-high electrical conductivity, a vast specific surface area, and superior mechanical properties—graphene has been extensively employed in studies focusing on the modification of zinc-ion batteries. This review will systematically introduce the multifaceted and multifunctional applications of graphene in aqueous zinc-ion batteries. People will focus on analyzing the mechanism of action and the modification effect of graphene when it is used as the protective layer of the negative electrode interface, the composite skeleton of the positive electrode, and the functional components of the electrolyte and separator. At the same time, researchers will focus on the latest technological breakthroughs of graphene materials in aqueous zinc-ion batteries, and ultimately point out the development challenges and directions of zinc-ion batteries. In the future, graphene can adjust the zinc affinity and ionic conductivity of batteries according to their charging and discharging states, achieving adaptive optimization of zinc dendrite behavior and providing a feasible foundation for the development of zinc-ion batteries with long service life and self-healing capabilities.

Keywords: graphene, zinc-ion battery, anode protection, cathode material

1. Introduction

Against the backdrop of the global energy transition towards cleanliness and low carbon, realizing the “dual carbon” goals involves developing safe, efficient, and large-scale energy storage technologies as an indispensable part. Although lithium-ion batteries are the market leaders in electrochemical energy storage, limitations including the rarity of lithium resources, elevated prices, and safety hazards of organic electrolytes have hindered their deployment in large-scale energy storage devices and other related areas [1,2]. Aqueous zinc-ion batteries (AZIBs) arose for their high specific capacity and low potential of zinc anodes, as well as the inherent safety of aqueous electrolytes, and have become a highly promising low-cost energy storage solution. However, in practical applications, AZIBs still faces many problems. Side reactions such as zinc dendrite growth

DOI: 10.54254/2755-2721/2026.BJ31522

at the negative electrode and hydrogen evolution corrosion seriously affect the battery's lifespan and safety. The electronic conductivity of the cathode transition metal oxide is low and its volume change is large, resulting in rapid capacity attenuation [3].

To address the aforementioned issues, methods such as electrode structure design and electrolyte optimization have been widely studied. Given graphene's high electron mobility, large specific surface area and superior mechanical properties, it has great application potential in AZIBs. In recent years, the research progress of graphene in AZIBs has been remarkable. Zhou et al. constructed a zinc anode functional interface layer using graphene nanoribbons, which increased the cycle life of symmetrical batteries by approximately 17 times [4]; Li et al. optimized the cathode performance through a molybdenum disulfide (MoS_2)/graphene heterostructure, achieving excellent rate performance [2]. This review aims to systematically summarize the application strategies and action mechanisms of graphene in AZIBs. This article will start with the protection of zinc anodes by graphene and the optimization of cathode materials, then explore its application in the functionalization of electrolytes and separators, and finally analyze current challenges and look forward to future directions, for the purpose of providing references for high-performance AZIBs research and development.

2. The application of graphene in anode protection and interface

2.1. The key issues faced by zinc anodes

The theoretical capacity of zinc metal anodes is high and their electric potential is low, but their practical application is severely restricted by three interrelated problems [4]. Firstly, because of the "tip effect" during the zinc deposition process, Zn^{2+} tends to deposit preferentially at the surface protrusions, causing the unconstrained growth of dendrites. These dendrites not only fall off the current collector, causing capacity attenuation, but also may pierce the separator, triggering battery short circuits [5]. Secondly, the deposition potential of zinc is much lower than the hydrogen evolution potential of water, which leads to competition between zinc deposition and hydrogen evolution reactions. Side reactions not only consume electrolytes and active substances but also generate hydrogen gas that raises the battery's internal pressure, posing a safety hazard [5]. Finally, zinc is thermodynamically unstable in aqueous environments and will spontaneously react with the electrolyte to form Zn4(OH)6SO4·nH2O and other passivation layers, increasing interfacial impedance, intensifying deposition inhomogeneity, and leading to a decrease in coulombic efficiency [4,5]. These issues jointly cause the technical bottleneck of short cycle life and poor reversibility of zinc anodes.

2.2. The mechanism of graphene as a protective layer for the two-dimensional interface layer

The construction of a graphene-based two-dimensional interface protective layer on zinc anode surface is direct and effective. Firstly, the dense graphene layer efficiently inhibits direct contact between water molecules and the zinc surface, thereby suppressing hydrogen evolution reactions and corrosion. Studies have shown that the graphene nanoribbon (GNRs) interface layer can significantly reduce the corrosion current density (icorr) from 0.83 mA cm^{-2} to 0.59 mA cm^{-2} [4]. Secondly, the high electrical conductivity of graphene can homogenize the electric field distribution on the electrode surface and guide the uniform deposition of Zn^{2+}. The layered reduced graphene oxide (rGO) formed by graphene oxide's (GO) self-driven reduction provides abundant zinc deposition nucleation sites, ensuring the uniform deposition of Zn^{2+} and enabling symmetrical cells

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to exhibit more stable voltage curves and smaller voltage hysteresis [4]. In addition, its excellent mechanical properties effectively suppress dendrite piercing.

3. The role of graphene in the structural design and modification of cathode materials

3.1. The mechanism and key issues of cathode materials

The energy storage mechanism of the cathode material for aqueous zinc-ion batteries mainly involves the reversible intercalation/deintercalation reaction of Zn^{2+} in the material lattice. However, this process is confronted with numerous problems. Firstly, most cathode materials (such as MnO_2, V_2O_5, MoS_2) are semiconductors or even insulators, resulting in slow charge transfer rates and poor rate performance [2,6]. Secondly, the intercalation/deintercalation of Zn^{2+} can cause significant volume changes, and repeated cycling leads to structural collapse and powdering of active substances [6,7]. In manganese-based materials, Mn^{3+} undergoes disproportionation reactions to form Mn^{2+} and dissolves in the electrolyte, resulting in irreversible loss of active substances and capacity attenuation [6,8]. In addition, strong electrostatic interaction between Zn^{2+} and the host material's lattice induces a high migration energy barrier and slow diffusion rate [6,7].

Li et al. successfully constructed a sandwich-like MoS_2/ graphene superlattice structure by ingeniously inserting rGO between MoS_2 layers through electrostatic adsorption-assisted hydrothermal methods [2]. This research aims to address the core issues of slow zinc ion migration kinetics and poor cycling stability caused by the narrow interlayer spacing and poor hydrophilicity of traditional MoS_2 cathode materials. It expands the interlayer spacing of MoS_2 from 0.62 nanometers to 1.16 nanometers, providing a spacious and rapid two-dimensional diffusion channel for larger hydrated zinc ions. Meanwhile, the intercalated graphene not only has excellent electronic conductivity itself, but also reduces its band gap to 0.34 eV, constructing an efficient charge transport network. The residual oxygen-containing functional groups on its surface enhance the composite's hydrophilicity and promote electrolyte infiltration. Thanks to this, the superlattice structure cathode material exhibits excellent electrochemical performance, maintaining high specific capacity at high current densities, with a capacity retention rate of 88.2% after 1800 cycles. However, the precise control of the intercalation amount of graphene in hydrothermal synthesis is quite challenging. Insufficient or excessive intercalation will affect the performance, and the reversibility of phase transformation and structural stability of this structure in long-term deep cycling still need further verification [2].

Cui et al. developed a high-quality supported cathode based on graphene-coated aluminum-vanadate (HAVO@G) nanoribbons. This research aims to address the core issues of poor electronic conductivity and slow ion transport of cathode materials under high-quality loads. The graphene coating constructs an efficient electron conduction path, which works in synergy with the large interlayer spacing of aluminum-vanadate to ensure that Zn^{2}+ can be rapidly intercalated and deintercalated even under a surface load of up to 15.69 mg cm^{-2}. At a current density of 2 A g^{-1}, the positive electrode achieved a specific capacity of 209.0 mAh g^{-1} and excellent cycling stability. However, such a high mass load may cause an increase in electrode thickness, influencing battery energy density, and the complex preparation process challenges large-scale production [8].

3.2. The multi-functional role of graphene in cathode materials

Graphene significantly improves the performance of the cathode materials of zinc-ion batteries through its unique structural advantages. Firstly, graphene sheets can closely wrap around the

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surface of the active material particles forming a large area “surface-to-surface” conductive contact. Compared with the “point-point” contact of traditional conductive agents, this may greatly reduce the interface resistance [2]; At the same time, these layers overlap with each other and form a three-dimensional conductive network, which intersects the entire electrode to improve the electronic conductivity. In addition to its ability to optimize conduction of electrons, its high specific surface area will ensure more contact area between electrolyte and electrode not only allowing relative number of electrochemical reaction-active sites but also inducing pseudocapacitive behavior which will be dominant to the charge storage process, ensuring better specific capacity and better rate performance of the electrode [9]. In terms of structural stability, the flexibility of graphene buffers the volume change of active substances during charge-discharge, which enhances structural stability. Moreover, the dense layer of a coating also provides a physical barrier to the dissolution and loss of active elements in the electrolyte, effectively improving the long-term cycling stability of the electrode [8]. For layered cathode materials, for instance, graphene can be used as a “pillar” to be placed between their layers, permanently widening the interlayer spacing. This structural modification directly decreases the migration energy barrier of zinc-ions, and provides favourable conditions for the fast intercalation and deintercalation of zinc-ions and the kinetics of ion diffusion [6]. Thus, by enhancing electrode electronic conductivity and ion diffusion capacity and structural stability, graphene provides a technical pathway to high-performance cathodes for zinc-ion batteries.

4. The functional application of graphene in electrolytes and diaphragms

4.1. Graphene is regulated by electrolyte addition to regulate the solvation structure and interface deposition of zn²+

In aqueous electrolytes, Zn^{2+} usually exists in the form of $[Zn(H_2O)_6]^{2+}$, and its high reactivity can trigger HER. When GO or GQD with abundant oxygen-containing functional groups are added, these additives have the ability to coordinate with the Zn^{2+}, substituted partially for the water molecules in its solvated sheath layer, to form a composite structure. This modification acts to diminish the activity and number of water molecules and in turn inhibits side reactions effectively. Among them, functional additives act in two key mechanisms synergistically to control uniform deposition of metallic zinc and effectively suppress the growth of dendrite. Additives of graphene oxide or graphene quantum dots can preferentially adsorb on the surface of zinc anode. The abundant functional groups on their surfaces are able to supply a large amount of zinc-philic sites, so as to greatly decrease the nucleation over potential of zinc, thus favoring the homogeneous nucleation of zinc ions [10]. In addition, some negatively charged additives, such as nitrogen and sulfur co-doped carbon quantum dots, were able to selectively enrich at zinc protrusions with uneven electric field distribution to form an electrostatic shielding layer. This effect can repel Zinc ions from depositing further towards the tip than it would otherwise - instead forcing the Zinc ions to preferentially deposit in areas with weaker electric fields. This contributes to reducing the ion concentration gradient at the electrode-electrolyte interface with benefits for the formation of a smooth and compact zinc deposit [11].

4.2. Graphene’s application as a diaphragm coating

Constructing a vertical graphene (VG) coating on the diaphragm surface can create a high specific surface area porous scaffold structure. Its conductive network can redistribute the interfacial electric field and guide the uniform transmission of Zn^{2+} flux, thus giving the nucleation and growth of

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dendrites at the interface a good avoidance. Li et al. created vertical graphene on the surface of commercial glass fiber diaphragms using the chemical vapor deposition technology. The three-dimensional conductive interface greatly reduced the local current density and got stable zinc deposition for more than 500 hours [1].

Meanwhile, the graphene coating greatly improves the mechanical strength and toughness of the diaphragm, physically preventing dendrite puncture. The tensile strength of the vertical graphene Janus separator prepared by Li et al. is more than three times that of traditional separators. This improvement of mechanical toughness can effectively resist the puncture force of the zinc dendrites during cycling process and avoid battery short circuits caused by separator damage [12].

5. Future outlook

To promote the transition of aqueous zinc-ion batteries from the laboratory to industrial application, future research needs to break through the limitations of single material optimization and shift towards collaborative systems engineering. The interface layer design should be upgraded from static protection to function-oriented, developing smart materials that can dynamically adjust the structure or zinc-philicity in response to current and potential, to achieve adaptive regulation that promotes uniform deposition during fast charging and enhances passivation protection during static storage.

The electrolyte system must be balanced in both high ionic conductivity and high mechanical strength. Gel or composite solid electrolytes should be developed for the expansion of voltage stability. In addition, the cathode material should build conductive network and ion conductive channels connected under high loads, and seek new mechanisms such as coordination reaction to break through the capacity limitations of traditional intercalation and deintercalation.

6. Conclusion

This paper systematically reviews the research progress of the application of graphene materials in AZIBs, providing a reference for solving the technical bottlenecks faced by AZIBs. To tackle the issues of dendrite growth, hydrogen evolution corrosion, and passivation layer formation on zinc anodes, the graphene-based two-dimensional interface protective layer enhances the anode's cycling stability by blocking water molecules, homogenizing the electric field distribution and inhibiting dendrite puncture. In cathode modification, graphene has improved the shortcomings of traditional cathode materials such as poor conductivity, easy structural collapse and slow ion diffusion through multiple effects such as constructing a three-dimensional conductive network, buffering volume changes and expanding the interlayer spacing, achieving a simultaneous increase in cycle lifespan and capability. In the field of electrolytes and separators, graphene-based additives have further suppressed side reactions and dendrite growth by regulating the solvation structure of Zn^{2+} and achieving uniform deposition, while separator coatings have optimized the electric field distribution and physical protection. In conclusion, graphene, with its ultra-high electrical conductivity, excellent mechanical properties and unique structural advantages, has exerted a crucial effect on the engineering of the zinc anode interface, the design of the cathode structure, and the functionalization of electrolytes and separators, providing technical support for the research and development of high-performance AZIBs. In the future, the focus should be on the practical application of graphene-based material optimization. By developing an intelligent responsive interface layer, the zinc-affinity and ionic conductivity of the battery can be adjusted according to its charging and discharging state, achieving further breakthroughs in the performance of AZIBs. At the same time, promote the large-

DOI: 10.54254/2755-2721/2026.BJ31522

scale production and cost control of graphene modification technology, and accelerate the commercial application of aqueous zinc-ion batteries in large-scale energy storage fields.

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