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# Stabilization Strategies of Carbon Materials for Zinc Anodes in High-Performance Zinc-Ion Batteries

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## Abstract

**With the escalating energy crisis and the booming development of wearable electronic devices, zinc-ion batteries (ZIBs) have emerged as a prominent research focus in electrochemical energy storage due to their excellent safety performance, low cost, and ecological compatibility. However, critical challenges such as dendritic zinc growth and parasitic byproduct formation at the zinc anode during cycling severely hinder their widespread commercialization. Carbon-based materials, distinguished by their exceptional electrical conductivity and intrinsic zincophilicity, have become a cornerstone in stabilizing high-performance zinc anodes. This review systematically summarizes the operational mechanisms of ZIBs, analyzes persistent challenges, evaluates carbon-material-based stabilization strategies , and finally proposes forward-looking perspectives on advanced carbon-engineered anodes for next-generation ZIBs.**

## Keywords

**Zinc-Ion Batteries; Zinc Anode Protection; Carbon Materials.**

## 1. Introduction

For a long time, exhaustible energy sources such as coal, oil, and natural gas have dominated global energy consumption, while recyclable energy sources like water and electricity account for only 11.41% of the total global energy consumption. The large-scale use of exhaustible energy sources has led to the current world facing both an energy crisis and an environmental crisis. There is an urgent need to transform the types of energy consumption [1]. Therefore, the use of recyclable energy sources such as wind, hydro, and tidal power has gradually increased in recent years. However, these energy sources are often influenced by climate conditions, characterized by intermittency and instability [2]. In contrast, electrochemical energy storage devices can convert these energy sources into chemical energy and release it when needed, thereby achieving stable energy supply. They are relatively efficient, environmentally friendly, and reliable. Meanwhile, as people's demands in areas such as fitness and health continue to grow, a variety of wearable and portable electronic devices are flourishing. Therefore, creating energy storage devices with superior performance, high safety, lightweight, and low cost to continuously power wearable electronics has become a current research hotspot [3]. Among traditional electrochemical energy storage devices, lithium-ion batteries stand out for their high energy density, high efficiency, and low self-discharge rate. They are most widely used in mobile electronic devices, transportation, and large-scale power grids [4]. However, several issues still hinder the further commercialization of lithium-ion batteries in the energy storage sector, such as the limited availability of lithium resources leading to increased production costs and safety concerns associated with organic electrolytes. Therefore, developing alternative battery technologies that are cost-effective and relatively safe holds significant importance [5]. In comparison, metallic

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zinc is abundant in reserves, with a high electrode potential (-0.76 V vs. standard hydrogen electrode, SHE), high specific capacity (5849 mAh $cm^{-3}$), lower production costs, environmental friendliness, and ease of processing. Consequently, aqueous zinc-ion batteries (ZIBs) have emerged as a promising alternative in the realm of electrochemical energy storage devices [6]. Unfortunately, during the operation of ZIBs, the zinc anode surface often faces the significant challenge of dendrite growth. This phenomenon is primarily caused by the uneven deposition of $Zn^{2+}$ on the anode surface, leading to a series of issues such as battery short circuits, shortened cycle life, and catastrophic capacity degradation. These problems greatly hinder the commercial application of ZIBs [7]. Therefore, overcoming the cycling degradation of zinc anodes is the core objective of current research.

Currently, various functional materials have been developed to address the aforementioned challenges, including metal oxides, carbon materials, metal-organic frameworks (MOFs), and MXenes. Among these materials, carbon-based materials have gained increasing attention due to their high chemical stability, excellent conductivity, and strong affinity for zinc. Many researchers have adopted the strategy of incorporating carbon materials to enhance the stability of zinc anodes, tackling a series of challenges during the cycling process of zinc anodes. For instance, leveraging the exceptional ability of carbon materials to regulate $Zn^{2+}$ deposition, graphene has been used as a protective coating for zinc foil. This promotes the preferential deposition of $Zn^{2+}$ onto the (002) plane during charging, inducing uniform zinc nucleation and significantly suppressing the formation of zinc dendrites and associated side reactions [8]. In addition to zinc foil itself serving as a current collector, traditional zinc anodes can also utilize materials such as copper foil and stainless steel foil as current collectors. However, these anodes are prone to galvanic corrosion during the cycling process, which significantly enhances hydrogen evolution reactions, leading to battery failure. This makes them unsuitable for long-term cycling in ZIBs [9]. In contrast, carbon-based materials such as graphene, carbon cloth, and carbon nanotubes exhibit superior corrosion inhibition capabilities when used as current collectors. Compared to metal current collectors, carbon materials are lighter in weight, which is more conducive to enhancing the energy density of batteries. Additionally, their lower interfacial transfer resistance and excellent thermal conductivity significantly improve the cycling and rate performance of the batteries [10]. In recent years, research on carbon-based materials as electrolyte additives has been flourishing. Carbon materials play a role in regulating the electric field distribution within the electrolyte, fundamentally addressing the uneven deposition and nucleation of $Zn^{2+}$. Additionally, they facilitate the desolvation process during $Zn^{2+}$ transport, significantly suppressing dendrite growth during battery cycling and stabilizing the zinc anode [8].

Therefore, as the role of carbon-based functional materials in strategies to stabilize practical zinc anodes continues to grow, their applications in areas such as anode interface layers and current collectors for ZIBs have become increasingly prevalent. It is essential to provide a brief introduction to several typical carbon materials, comprehensively summarize their various strategies for enhancing zinc anode stability, and analyze their specific regulatory mechanisms during the cycling process. This will offer new insights for the development of next-generation ZIBs. This article first introduces the energy storage mechanisms of ZIBs and briefly outlines the challenges encountered during their cycling. It then reviews the latest strategies and design approaches using different carbon materials (such as graphene, carbon nanotubes, and carbon fibers) to address these issues. Finally, the article discusses and prospects the future practical applications of carbon materials in the field of ZIBs energy storage.

## 2. Energy Storage Mechanisms and Challenges of ZIBs

ZIBs are primarily composed of four components: a Zn metal anode, a cathode with tunnel or layered structures, an electrolyte containing $Zn^{2+}$, and a separator. Among these, the anode provides reversible $Zn^{2+}$/Zn conversion, the cathode acts as a host for $Zn^{2+}$ during energy storage, the electrolyte serves as the pathway for zinc ions to migrate from the anode to the cathode, and the separator acts as a barrier between the anode and the cathode, thereby avoiding short circuits and thermal runaway. Generally, the anode material in ZIBs can only be Zn metal as the active material, while the cathode

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materials mainly include four types: manganese-based oxides, vanadium-based oxides or vanadates, and Prussian blue analogs. Unlike anode materials, almost every cathode material has its unique reaction characteristics, with reversible $Zn^{2+}$ intercalation and deintercalation being the most common energy storage mechanism. Taking a ZIBs with Zn as the anode and $MnO_2$ as the cathode as an example, during discharge, the Zn anode loses electrons to form $Zn^{2+}$, which then gains electrons in the electrolyte and embeds into the cathode. At this stage, $ZnMn_2O_4$ is generated on the cathode surface. During reverse charging, $Zn^{2+}$ is redeposited onto the anode surface to form Zn metal. The specific chemical reactions occurring at the anode and cathode are shown in Equations(1,2)[11, 12].

$$Zn \leftrightarrow Zn2^{+} + 2e^{-} \tag{1}$$

$$Zn^{2+} + 2e^{-} + 2MnO_2 \leftrightarrow ZnMn_2O_4 \tag{2}$$

Although ZIBs offer numerous advantages such as high stability, long cycle life, and environmental friendliness, they still face various challenges during use. Firstly, needle-like protrusions often appear on the anode surface during the cycling of ZIBs, a phenomenon known as zinc dendrites. This occurs due to the unstable deposition of $Zn^{2+}$ migrating from the cathode to the anode surface during the charging process. Specifically, the deposition of $Zn^{2+}$ involves both nucleation and growth of zinc, processes that are significantly influenced by the electric field and ion distribution on the anode surface. Electrons and ions tend to migrate to regions with enhanced electric fields and high zinc concentrations. Subsequently, zinc nucleation spontaneously grows on existing protrusions due to its high surface energy. The intensified electric field and increasingly concentrated $Zn^{2+}$ further promote the nucleation and growth of zinc metal in specific areas, gradually evolving into zinc dendrites. As the number of battery cycles increases, the formation of dendrites raises the surface roughness, leading to the continuous growth of needle-like rough zinc dendrites. The continuous evolution of zinc dendrites will cause mechanical failure of the separator, leading to ohmic contact between the electrodes and triggering an internal short - circuit. This can result in battery swelling and ignition, severely impacting the normal use of the battery. Moreover, the hydrogen evolution reaction that occurs when zinc comes into contact with water in the electrolyte can also severely impact its service life. The water in the electrolyte gains electrons to produce hydrogen gas, which accumulates on the surface of the anode. Simultaneously, the enriched $OH^-$ on the anode surface facilitates the formation of by-products such as zinc oxide and basic zinc sulfate, leading to the creation of a significant amount of "dead zinc." This greatly hinders the normal plating and stripping of $Zn^{2+}$. The formation of insoluble by-products also increases the internal resistance of the electrode surface and affects the distribution of the electric field on the anode surface, promoting the growth of dendrites. The vertical growth of dendrites increases the active area of the zinc anode exposed to the electrolyte, which in turn further accelerates the hydrogen evolution reaction [6]. Therefore, it can be said that there exists a mutually reinforcing relationship between dendrite growth and the hydrogen evolution reaction. These phenomena have significantly restricted the industrialization process of ZIBs. To address the aforementioned issues, extensive research has been conducted on the introduction of carbon materials to inhibit these reactions. Following this, the characteristics of three types of carbon materials (graphene, carbon nanotubes, and carbon fibers) will be introduced, and a detailed review of their strategies in stabilizing zinc anodes will be provided.

## 3. Graphene

The growth modes of zinc crystals on the anode surface primarily include three types: horizontal alignment, vertical growth, and random arrangement. When stainless steel is used as the substrate template for zinc crystal growth, zinc deposition predominantly exhibits random nucleation and vertical alignment on the surface. The reversibility of zinc deposition/dissolution is strongly correlated with the crystallographic orientation of zinc crystals: vertically oriented growth

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(perpendicular to the substrate) promotes the formation of zinc dendrites due to localized electric field enhancement, leading to irreversible capacity loss through internal short circuits and active material detachment. Therefore, inducing planar-oriented zinc crystal growth has been demonstrated to suppress dendrite propagation and significantly extend the cycle life of ZIBs.

As a single-layer two-dimensional planar material, graphene has excellent properties such as high electrical conductivity, large specific surface area, and high chemical stability.Its excellent charge carrier mobility and low lattice defects can significantly enhance the deposition kinetics of zinc, facilitating dendrite-free zinc deposition [13]. For instance, when a layer of graphene is coated on the surface of stainless steel, the growth direction of zinc crystals during the subsequent deposition process often exhibits a horizontal alignment rather than random arrangement or vertical growth. This indicates that a graphene interface with low lattice defects can regulate the growth direction of zinc crystals, which is evidently beneficial for dendrite-free deposition on the zinc anode surface. Therefore, the deposition of uniform, dense, and parallel-aligned zinc nanosheets on the graphene interface is more conducive to enhancing the long-term cycling performance of the zinc anode under high-rate conditions [14]. Based on this, Xia et al. utilized the spontaneous redox reaction between graphene oxide (GO) and zinc foil to self-assemble a layered reduced graphene oxide (rGO) film on the surface of the zinc foil [15]. The Zn/rGO hybrid zinc anode regulates the electric field distribution on the anode surface, providing a larger plating/stripping surface area and more nucleation sites, which encourages $Zn^{2+}$ to deposit horizontally, effectively limiting the formation of zinc dendrites. After 250 cycles at 0.2 mA $cm^{-2}$, the anode maintains a relatively smooth morphology with no significant dendrite formation. It can sustain a cycling time of up to 300 hours at 1 mA $cm^{-2}$. The zinc-ion hybrid supercapacitor assembled with this anode exhibits outstanding cycling performance, retaining 88.5% of its capacity after 5000 cycles. Coupled with its excellent rate performance, this demonstrates the broad application prospects of the Zn/rGO anode for enhancing the electrochemical performance of devices.

Self-assembling two-dimensional graphene materials within zinc foam to form 2D nanosheet-interpenetrated zinc foil anodes is also an effective method for preparing high-performance zinc anodes. For instance, Li et al. self-assembled rGO nanosheets inside a zinc foam skeleton and used mechanical compression to fabricate a hybrid zinc anode (GiZn) [16]. The excellent hydrophilicity and conductivity of rGO are beneficial for enhancing the ionic/electronic conductivity of the zinc anode, while its superior zincophilicity also helps regulate the uniform deposition of $Zn^{2+}$. Additionally, the porous structure in GiZn can accommodate more $Zn^{2+}$, enabling the zinc anode to withstand higher deposition capacities. Symmetric cells assembled based on this anode have been cycled for over 790 hours at 0.5 mA $cm^{-2}$ and for more than 300 hours at 10 mA $cm^{-2}$. The assembled GiZn||$MnO_2$ full cell also demonstrates a high capacity retention rate of 77% at 2C, proving the practical performance of 2D nanosheet-interpenetrated Zn hybrid foil for advanced ZIBs. The low utilization rate of zinc anodes is another critical issue in the development of ZIBs, as it significantly limits the energy density of the battery. To address this, Lin et al. prepared a binder-free, self-supporting 3D Zn_G anode using zinc powder and graphene oxide [4]. This anode boasts high utilization and a low N/P ratio, among other advantages. The graphene sheets enveloping the zinc powder surface effectively curb corrosion and side reactions, significantly reducing dendrite formation. The symmetric battery assembled with the Zn_G anode has a cycle life exceeding 570 hours at a current density of 1 mA $cm^{-2}$, with a low voltage hysteresis of only 20 mV. When paired with a $MnO_2$ cathode, the full cell achieves up to 1600 cycles at a current density of 0.5 A $g^{-1}$, with a specific cycle capacity as high as 131 mAh $g^{-1}$, offering greater potential for the development of high-performance zinc powder anodes.

## 4. Carbon Nanotubes

Carbon nanotubes (CNTs), leveraging their high conductivity, structural tunability, and low $Zn^{2+}$ nucleation barrier, serve as an ideal host material for zinc anodes: The constructed 3D conductive network not only homogenizes the electric field distribution for $Zn^{2+}$ deposition but also accelerates

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deposition/dissolution reaction kinetics, thereby effectively suppressing the uncontrolled growth of zinc dendrites through spatial confinement and interfacial charge redistribution mechanisms [17].

Zhang et al. utilized the high electronic conductivity of carbon nanotubes (CNTs) by combining them with ethylene-vinyl acetate (EVA) and zinc powder to fabricate a ZnP-MIEC scaffold with high ionic and electronic conductivity [18]. The zinc powder coated with EVA effectively avoids corrosion in the electrolyte. Electrochemical tests reveal that this hybrid anode exhibits excellent performance in suppressing hydrogen evolution and corrosion. Compared to zinc foil, it has a lower interfacial transfer resistance, approximately 1036 Ω. When assembled into a symmetric battery, the hybrid anode demonstrates a cycle life exceeding 1200 hours at 0.25 mA $cm^{-2}$ and remains stable after 100 hours of cycling at a high current density of 10 mA $cm^{-2}$, significantly outperforming zinc foil. Through characterization techniques such as SEM and XRD, the ZnP-MIEC anode shows superior performance in suppressing dendrite formation and by-products compared to zinc foil. In full-cell electrochemical testing, the full battery assembled with the ZnP-MIEC scaffold exhibits outstanding capacity performance at both high and low rates, demonstrating its excellent ion storage capability under high current densities. At 0.2 A $g^{-1}$, it achieves over 1600 cycles with a capacity retention rate exceeding 90%. When connected in series, it successfully powers an LED, demonstrating their great potential as electrochemical energy storage devices.

Inspired by the excellent regulatory effect of carbon nanotubes (CNTs) on $Zn^{2+}$ deposition, Zeng et al. utilized chemical vapor deposition (CVD) to grow CNT arrays and zinc nanosheets on carbon cloth, fabricating a Zn/CNT electrode [17]. Compared to bare carbon cloth, the zinc nanosheets on the surface of the Zn/CNT electrode appear more uniform and dense. The assembled symmetric battery can cycle for 200 hours at 2 mA $cm^{-2}$ with a polarization voltage of only 27 mV, whereas the Zn/CC electrode symmetric battery shows abnormal voltage distribution after just 50 hours of cycling. The Zn/CNT electrode symmetric battery exhibits a cycle life exceeding 110 hours at 5 mA $cm^{-2}$, far surpassing the 30 hours of the control sample. This demonstrates the excellent cycling performance of the Zn/CNT symmetric battery at different current densities. To evaluate the practical application performance of the Zn/CNT electrode, it was assembled into a Zn/CNT||$MnO_2$ full battery. According to CV test results, the full battery with deposited CNTs shows smaller voltage polarization, corresponding to its lower interfacial charge transfer resistance. In terms of cycling performance, the Zn/CNT||$MnO_2$ full battery can cycle for over 1000 cycles at 2 mA $cm^{-2}$ with a capacity retention rate close to 90%, higher than the 69.3% of the Zn||$MnO_2$ battery. This highlights the outstanding performance of CNTs in enhancing the durability of zinc anodes.

Constructing an artificial conductive coating is a practical method for preparing high-performance zinc anodes. This process helps balance the ion distribution on the anode and ensures uniform $Zn^{2+}$ deposition. Cho et al. employed a simple coating method to create a micron-thick layer of single-walled carbon nanotubes (SWCNTs) on the surface of copper foil [19]. The CNT@Cu||Zn half-cell achieves a stable cycle life of 2900 hours at 1 mA $cm^{-2}$, and after 1500 cycles at current densities ranging from 0.1 to 10 mA $cm^{-2}$, it maintains an excellent Coulombic efficiency close to 100%. In terms of full - cell applications, the CNT@Cu electrode can deliver an outstanding capacity exceeding 300 mAh $g^{-1}$. Its excellent performance in rate capability tests further demonstrates the remarkable stability of the single - walled carbon nanotube layer, providing new ideas for the future development of modifying zinc anodes with copper current collectors.

## 5. Carbon Fiber

Carbon fiber is not only lightweight but also exhibits excellent conductivity and superior mechanical properties, making it a common material used in energy storage devices such as batteries and supercapacitors. Generally, it can be directly used as an anode or as a current collector material. However, when used directly as an anode, the volume changes caused by ion deposition can limit the electrode's lifespan. When employed as a current collector, the electrode structure often struggles to maintain integrity under external forces. Consequently, significant efforts have been made to enhance

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its performance in zinc anode applications [20]. For example, Ying and colleagues constructed a multi-chambered carbon fiber (MCF) interfacial layer on the surface of zinc foil to enhance the stability of the zinc anode. The MCF exhibits high electrolyte wettability while also improving the distribution of the electric field on the anode surface [21]. Through characterization and computational analysis, it has been determined that the MCF interface layer exhibits excellent zincophilic properties, enabling bottom-up growth of zinc. In terms of symmetric battery cycling performance, even under a high areal capacity of 5 mAh $cm^{-2}$, the MCF interface layer can still provide a cycle life exceeding 1600 hours. The assembled Cu@MCFs||Zn half-cell demonstrates a Coulombic efficiency close to 100%. To further validate the practical application of the MCF interface layer, the assembled pouch cell maintains a capacity retention rate of 82.93% after 250 cycles at 2 A $g^{-1}$, proving the outstanding capacity performance of the Zn@MCFs anode. Yang et al. constructed a 3D Sn-PCF modified scaffold as a host for zinc deposition [22]. The synergistic effect between highly conductive carbon fibers and surface-modified Sn-philic coatings reduces charge transfer impedance and induces epitaxial growth. Compared to a Cu substrate, the 3D Sn-PCF host scaffold exhibits a flat and dense surface after zinc deposition, with no significant zinc dendrite formation. At a current density of 1 mA $cm^{-2}$, the symmetric battery assembled with the Sn-PCF@Zn anode demonstrates an ultra-long cycle life (750 hours) and a low polarization voltage (22 mV). Even at a high current density of 10 mA $cm^{-2}$, it still exhibits excellent cycling stability. In the extended application of the 3D Sn-PCF scaffold, the Zn||NaVO full cell demonstrates exceptional capacity performance and an ultra-long cycle life at 10 A $g^{-1}$, maintaining a capacity retention rate of 73.5% after 2500 cycles. This provides a new design approach for element doping and modification of carbon materials.

## 6. Conclusion

In recent years, with the ever - increasing global demand for sustainable energy storage solutions, zinc - ion batteries (ZIBs) have been regarded as highly promising functional units for electrochemical energy storage devices, owing to their remarkable advantages such as high safety, excellent ecological compatibility, and low cost. Nevertheless, issues such as dendrite growth on the surface of zinc anodes and the accompanying side reactions still remain the major obstacles hindering their large - scale practical applications. This paper, focusing on stabilizing the zinc anode, outlines the fundamental properties of three common carbon materials—graphene, carbon nanotubes, and carbon fibers-and their various practical strategies in zinc anode applications. These three carbon materials can effectively improve electric field distribution, enhance conductivity, suppress the "tip effect," and inhibit dendrite growth. Among several typical strategies, carbon materials, whether used as current collectors, interfacial layers for zinc anodes, or highly zincophilic substrates, have successfully achieved uniform and stable $Zn^{2+}$ deposition as well as highly reversible plating/stripping processes. However, to further expand their use in stabilizing ZIBs anodes, certain difficulties and challenges still need to be addressed. Here, we offer some perspectives and insights on the role of carbon materials in stabilizing zinc anodes. First, during the preparation of carbon-based composite zinc anodes, the ratio of carbon materials to active substances must be strictly controlled. Excessive carbon content can reduce the energy density of zinc-ion batteries, making it crucial to achieve the highest electrochemical performance with limited carbon material usage. Second, when carbon materials are used as protective coatings for zinc anodes, the volume changes caused by continuous $Zn^{2+}$ plating/stripping can weaken the contact between the interfacial layer and the substrate. Enhancing the anchoring effect of the substrate on the interfacial layer to accommodate long-term $Zn^{2+}$ deposition is an urgent issue to resolve. Third, in addition to mixing carbon materials with zinc to prepare anodes, incorporating other highly zincophilic elements such as Se, Te, and Bi to create hybrid anodes could further improve anode stability. Fourth, current processing methods for carbon-material hybrid zinc anodes still face challenges such as high costs and difficulties in large-scale production, which hinder practical manufacturing needs. Developing simple, practical, and scalable processing methods for zinc anodes is essential for future energy storage systems.

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