Research Progress on High-Performance Current Collectors for Zinc-Ion Hybrid Supercapacitors

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Abstract

Zinc-ion hybrid supercapacitors (ZIHSs), integrating the high power density of supercapacitors with the high energy density of zinc-ion batteries, have emerged as a cutting-edge technology in advanced energy storage systems. As a critical component bridging electrodes and external circuits, current collectors directly govern the electrical conductivity, interfacial stability, and cycling longevity of ZIHS devices. Recent efforts have focused on optimizing current collectors through material selection, structural design, and surface modification to address performance limitations. This review systematically outlines the fundamental requirements for ZIHS current collectors, critically evaluates recent advancements in metal-based, carbon-based, and composite current collectors, and provides insights into future research directions for developing high-performance architectures. Furthermore, challenges and opportunities in scalable fabrication, low-temperature adaptability, and intelligent functionality integration are discussed to guide the rational design of next-generation ZIHS systems for practical applications.

Keywords

Energy Storage; Zinc-ion Hybrid Supercapacitors; Current Collector.

1. Introduction

The escalating energy and climate crises caused by excessive fossil fuel exploitation have heightened global awareness of the imperative to develop renewable energy systems. Consequently, the proportion of clean energy sources such as wind, hydro, and solar power has grown substantially in recent years. However, their inherent intermittency and fluctuations pose challenges to stable utilization, driving urgent demands for efficient energy storage technologies [1]. Flexible, high-performance electrochemical energy storage systems have thus emerged as critical solutions. Since the 21st century, lithium-ion batteries (LIBs) have dominated applications in electric vehicles and wearable electronics due to their high energy density and long cycle life. Nevertheless, limitations including lithium resource scarcity, safety concerns, cost pressures, and low power density have spurred exploration of alternative technologies [2]. Supercapacitors, relying on physical charge adsorption mechanisms, complement batteries with ultrahigh power density but suffer from limited energy density, restricting their broad adoption. Developing hybrid systems that synergize high energy and power densities remains a pivotal goal [3].

Recent advancements in zinc-ion hybrid supercapacitors (ZIHSs), which combine battery-level energy density with capacitor-like power density, demonstrate promising potential for grid peak shaving and distributed energy storage [4]. The practical performance of ZIHSs, however, is often constrained by electrode stability, where rational electrode design is crucial for commercialization.

As a core component, current collectors serve as electronic pathways between active materials and external circuits, ensuring rapid electron transfer during charge/discharge cycles while minimizing interfacial resistance. They also provide robust physical support to maintain uniform loading and prevent active material detachment, particularly critical for preserving electrode integrity during prolonged cycling. Consequently, current collectors directly dictate device energy density, cycling stability, and rate capability. Existing materials still face challenges such as insufficient conductivity, interfacial instability, high costs, and electrochemical corrosion in zinc-ion electrolytes [5]. Despite extensive research on active materials and electrolytes, systematic reviews categorizing ZIHS current collectors remain scarce. This work addresses this gap by outlining critical performance requirements for ZIHS current collectors, analyzing three mainstream material categories (metal-based, carbon-based, and composite architectures), and discussing their optimization strategies. Finally, we summarize key challenges, opportunities, and future directions to guide the rational design of advanced current collectors for high-performance ZIHS systems.

2. Functional Requirements for ZIHS Current Collectors

The electrochemical performance of zinc-ion hybrid supercapacitors (ZIHSs), including energy density, power density, and cycling stability, is synergistically governed by their core components-active materials, current collectors, electrolytes, and separators. Optimizing the material properties and structural design of each component is critical for enhancing overall device performance and advancing practical applications [6]. As a pivotal interface between electrode materials and external circuits, current collectors directly dictate electrical conductivity, cycling durability, and energy efficiency. Ideal current collectors must exhibit high interfacial stability with electrodes, robust chemical inertness in electrolytes, superior mechanical strength, high modulus, excellent conductivity, and cost-effectiveness [7]. Specifically, ZIHS current collectors require high electrical conductivity to ensure efficient electron transport, as well as exceptional mechanical robustness to withstand volume fluctuations and mechanical stresses induced by repeated Zn^{2+} intercalation/deintercalation or deposition/dissolution during cycling. Electrochemical stability is essential to suppress parasitic reactionsin ZIHS electrolytes, while strong interfacial compatibility with electrode materials ensures stable interfaces and efficient ion/electron transport. Lightweight design is equally critical to minimize inactive mass and improve gravimetric energy density, with low-density carbon-based materials being favored for wearable electronics due to their optimal conductivity-to-weight ratios [8]. Additionally, cost-effective fabrication and processability are imperative to enable scalable production and complex structural designs for performance enhancement. Addressing these requirements holistically will accelerate ZIHS commercialization by balancing performance, durability, and economic feasibility.

3. Classification and Recent Progress of Current Collector Materials

3.1 Metal-Based Current Collector

In recent years, metallic current collectors with high conductivity and mechanical strength, such as nickel, copper, aluminum (noted for conductivity), and titanium or stainless steel (valued for chemical stability and robustness), have attracted significant attention and are widely employed in zinc-ion hybrid supercapacitors (ZIHSs) due to their superior properties [9]. However, challenges remain during ZIHS cycling. For instance, in aqueous electrolytes (e.g., $ZnSO_4$ or $Zn(CF_3SO_3)_2$, metallic current collectors are susceptible to corrosion and side reactions, leading to collector failure and depletion of active electrolyte components, thereby degrading device performance. Additionally, interfacial compatibility and contact resistance between metallic collectors and electrode materials require optimization to enhance power density and cycling stability. Common modification strategies include coating conductive layers (e.g., carbon, graphene, conductive polymers) or protective layers on metallic surfaces. For example, Xi et al. utilized a shear-flow strategy to fabricate horizontally aligned graphene-coated copper current collectors (Cu@G) at scale [10]. In battery-type anodes, the Cu@G collector enabled denser and more uniform zinc deposition compared to bare Cu, effectively suppressing dendrite growth. The Cu@G@Zn electrode exhibited higher hydrogen evolution overpotential, reduced corrosion current, and shifted corrosion potential, confirming enhanced corrosion and side reaction resistance. When paired with an activated carbon (AC) cathode, the assembled ZIHS demonstrated quasi-rectangular cyclic voltammetry (CV) profiles across scan rates and achieved over 45,000 cycles at 40 mA cm^{-2} with a high areal capacity (2 mAh cm^{-2}) and ~98% Coulombic efficiency, underscoring the potential of carbon coatings for improving metallic collectors.

Composite approaches, such as integrating metals with MXene via alkalization and pre-intercalation to form 3D ribbon-like AMX-Zn anodes (functioning as dual-purpose current collectors), have also been explored [11]. Devices assembled with porous carbon (PC) cathodes retained stable CV curves even at 100 mV s^{-1} and displayed highly reversible charge/discharge profiles across current densities, maintaining ~93% capacity after 10,000 cycles. Interfacial engineering further addresses contact issues. Zhang et al. engineered a 3D porous alumina structure on aluminum foil via anodic oxidation, followed by carbon coating via CVD, yielding a carbon/alumina/aluminum (CAAO) composite film [12]. The MoO_{3-x}/PPy@CAAO anode exhibited an areal capacitance of ~1400 mF cm^{-2}, and the assembled device with a V-MnO_2@CAAO cathode achieved an areal energy density of 140.9 μWh cm^{-2} and power density of 0.9 mW cm^{-2}. The CAAO collector retained >90% capacitance under bending angles of 0–180°, highlighting its flexibility and potential for advanced 3D carbon-metal composite designs.

3.2 Carbon-Based Current Collector

Although existing modification strategies for metallic current collectors can partially mitigate corrosion and side reactions, inherent limitations persist. For instance, the high density and weight of metals conflict with the lightweight demands of modern electronics, while their rigid structures and susceptibility to cyclic stress failure hinder compatibility with flexible devices. Additionally, zinc dendrite formation on metallic collectors serving as anodes severely restricts device lifespan [13]. In contrast, carbon-based materials such as graphene, carbon nanotubes, and carbon cloth have emerged as promising alternatives due to their lightweight nature, high conductivity, and large specific surface area [14]. However, challenges including poor electrolyte wettability, high zinc nucleation overpotential (>100 mV on unmodified carbon cloth), and low mechanical strength hinder their practical application. To address these issues, various modification strategies have been explored. For example, Jia et al. employed oxygen plasma treatment to functionalize carbon cloth (OPCC) as a ZIHS current collector [15]. The OPCC, enriched with oxygen-containing groups, regulated ion/electrical field distribution, enhanced zinc nucleation kinetics, and improved electrolyte wettability, resulting in more uniform zinc deposition compared to untreated carbon cloth. When paired with an activated carbon cathode, the assembled ZIHS exhibited exceptional cycling stability (99% capacity retention after 1,500 cycles at 1 A g^{-1}) and rate capability, outperforming devices with untreated carbon cloth across current densities (0.1–5 A g^{-1}). Moreover, the device maintained its excellent performance even under extreme bending conditions, demonstrating its potential applications in flexible energy storage systems.

Laser-induced graphene (LIG)-based micro-supercapacitors (LIG-MSCs) have advanced rapidly, yet their energy density remains limited by the low specific surface area of commercial polyimide-derived LIG current collectors. To overcome this, Dong et al. developed fluorinated polyimide-derived porous fluorinated LIG (FLIG) via laser irradiation, significantly enhancing specific surface area [16]. Zinc electrodeposited on FLIG exhibited uniform and dense morphology, enabling a device with elevated energy density (15.1 μWh cm^{-2}). The FLIG-based device demonstrated a 15-fold increase in areal capacitance (42.32 mF cm^{-2}) compared to commercial counterparts, along with 7,000 cycles at 1 mA cm^{-2} (80% capacity retention), showcasing promise for wearable electronics. Three-dimensional graphene frameworks offer high conductivity and surface area but face scalability challenges due to complex fabrication. Kwon et al. proposed a cost-effective, scalable method to produce graphite-graphene hybrid current collectors (GF) that simultaneously function as active materials [17]. The oxygen-functionalized GF delivered an areal capacitance of 140 mF cm^{-2} at 1 mA cm^{-2}, retaining 84 mF cm^{-2} at 10 mA cm^{-2}, underscoring its potential for mass production of high-performance ZIHS systems.

3.3 Hybrid Current Collector

In addition to metallic and carbon-based current collectors, composite architectures such as metal-carbon and polymer-carbon hybrids leverage multi-material synergies to enhance the energy density, cycling longevity, and environmental adaptability of zinc-ion hybrid supercapacitors (ZIHSs). For instance, replacing traditional zinc anodes with pseudocapacitive alternatives has gained traction to mitigate dendrite growth and side reactions. Ji et al. developed a polyimide-grown multi-walled carbon nanotube (PNDIE/MWCNT) composite anode, which simultaneously functions as both active material and current collector [18]. By circumventing zinc dendrite formation, the ZIHS assembled with a PEDOT-Na0.55MnO2/CC cathode retained over 80% capacity after 2,000 cycles. Owing to the lightweight carbon framework, the device achieved a high energy density of 10.7 Wh kg^{-1} and a power density approaching 200 W kg^{-1}, offering a promising pathway for non-metallic anodes. Similarly, Hung et al. coated copper foil with a ZIF-8/sodium alginate (SA) hybrid (Z8-SA@Cu) as a dual-purpose current collector and active material [19]. The Z8-SA@Cu anode exhibited lower zinc nucleation barriers and superior suppression of parasitic reactions compared to zinc foil. The assembled device demonstrated stable capacity retention (~90% after 12,000 cycles at 5 mA) across current densities.

In another strategy addressing dendrites and side reactions, Jiang et al. integrated carbon cloth with gallium-doped liquid metal (LM) as a zinc anode current collector [20]. This design enhanced anode flexibility and electrolyte wettability, enabling a ZIHS with an activated carbon (AC) cathode to deliver a specific capacity of 65 mAh·g^{-1} at 2 A g^{-1}, along with 95% capacity retention after extended cycling (40% longer lifespan than conventional counterparts). The device maintained its performance under mechanical bending and successfully powers LED lights, underscoring its potential for flexible energy storage applications.

4. Conclusion

With the continuous growth of demand for high-performance energy storage in electronic devices, research on zinc ion hybrid supercapacitors has attracted much attention. As the core component of ZIHS, the material selection and structural design of the current collector play a decisive role in device performance: surface functionalization of carbon-based materials, integrated design of metal-based materials, and synergistic effects of composite systems have effectively improved energy density and cycle life. However, current research still faces some key challenges and opportunities. If current collectors face bottlenecks in large-scale preparation, existing modified current collectors rely heavily on complex processes. In the future, it is necessary to explore low-cost and batch production technologies to promote the industrialization process of high-performance current collectors. Secondly, the adaptability of current collectors to low temperature environments is insufficient. Traditional current collectors suffer from severe performance degradation at low temperatures due to electrolyte solidification and increased interfacial impedance. A synergistic system of antifreeze electrolyte and low-temperature resistant current collector needs to be developed to overcome the application limitations of extreme environments. Finally, the dynamic self-healing function of the current collector is lacking, and current research mainly focuses on passive inhibition of zinc dendrites and side reactions, with insufficient ability for dynamic regulation. Developing intelligent current collectors with self-healing capabilities is expected to achieve in-situ dendrite elimination, thereby significantly improving cycle stability. In summary, breaking through the three major bottlenecks of fluid collection preparation technology, environmental adaptability, and functional intelligence will promote the widespread application of ZIHS in flexible electronics, extreme environment energy storage, and wearable devices. Future research needs to integrate material innovation, structural design, and interdisciplinary technology to achieve the dual goals of performance optimization and practical implementation.

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