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Reassessing the Commercialization of Aqueous Zinc-Ion Batteries Based on Laboratory Achievements

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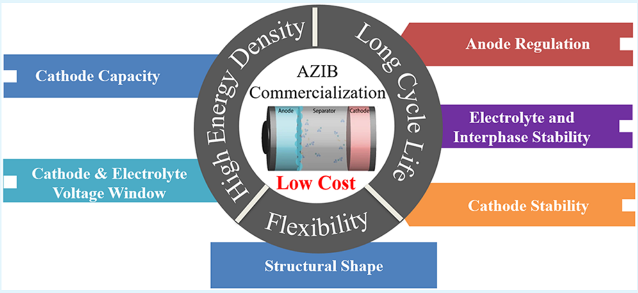

ABSTRACT: The development of aqueous zinc-ion batteries (AZIBs) has attracted increasing attention as a promising route toward low-cost, safe, and sustainable energy storage. While their intrinsic advantages, such as nonflammable electrolytes, abundant zinc resources, and environmentally benign chemistries, position AZIBs as attractive candidates for long-lifespan electrochemical energy storage, significant technical gaps remain between current laboratory-scale innovations and system-level deployment requirements. This review analyzes recent advancements in cathode materials, electrolyte formulations, and interfacial stabilization strategies, with a focus on how these approaches impact performance metrics relevant to practical applications. Drawing from both peer-reviewed studies and publicly available data from the U.S. Department of Energy, we derive target performance thresholds and use these benchmarks to assess the readiness of key material systems including manganese- and vanadium-based cathodes. Particular emphasis is placed on strategies such as defect engineering, water intercalation, and electrolyte-electrode compatibility for enhancing energy density and cycle stability. Despite the notable progress, challenges such as structural degradation, transition metal dissolution, and limited Zn^{2+} transport kinetics greatly impede the commercial viability of AZIBs. By bridging insights from laboratory results with projected deployment criteria, this review provides a performance-oriented roadmap to guide future research efforts aimed at transforming AZIBs from proof-of-concept systems to commercialized technologies.

KEYWORDS: *aqueous zinc-ion batteries, commercialization, electrolyte optimization, cathode dissolution, separator design, current collector engineering, structural battery composites*

1. INTRODUCTION

With growing efforts to expand renewable energy and reduce fossil fuel dependence, the importance of large-scale energy storage systems (EESs) is rising.[1] Lithium-ion batteries (LIBs) have been widely used due to their high energy density.[1−3] However, the LIB limitations, such as scarce lithium resources, high costs, and safety concerns, have led to the development of alternative EESs.[4] As a promising candidate for next-generation energy storage, aqueous zinc-ion batteries (AZIBs) have gained great attention due to their low cost, excellent safety, and eco-friendliness.[5,6] Particularly, zinc is approximately 300 times more abundant than lithium, and zinc metal (\$3.5 kg^{-1}) is significantly cheaper than lithium (\$19 kg^{-1}) (Table 1). AZIBs also use noncombustible aqueous electrolytes that provide high safety, whereas conventional LIBs utilize highly flammable organic electrolytes, which lead to potential risks of fire and explosion.[7,8] However, challenges hindering AZIB commercialization include low energy density (currently below 135 Wh kg^{-1}), hydrogen evolution at the zinc anode, and electrolyte instability during charge/discharge cycles.

To meet the U.S. Department of Energy's (DOE) long-duration energy storage (LDES) target of achieving a levelized cost of storage (LCOS) below \$0.05 kWh^{-1}, zinc-based batteries, particularly AZIBs, must overcome several key performance limitations. Current projections for 2030 estimate AZIB systems to reach an LCOS of approximately \$0.05 kWh^{-1}, with a cycle life of around 6500 cycles, a calendar life of 17 years, and a round-trip efficiency (RTE) of approximately 74% (Table 2).[14] However, further improvements in both energy density and cycle life are essential to fully meet these targets. According to the technology strategy assessment of DOE, advances in zinc metal anode design and cathode material optimization could improve cycle life by more than

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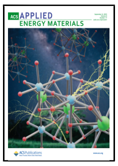

Table 1. Comparison of Aqueous Zinc-Ion Batteries (AZIBs) and Li-Ion Batteries (LIBs)[a,9−16]

	AZIBs	LIBs
anode	Zn	graphite
cathode	MnO_2, VO_x, etc.	NMC, LFP, NCA, LCO
electrolyte	aqueous (e.g., $ZnSO_4$)	organic solvents (e.g., EC/DMC with $LiPF_6$)
working voltage (V)	∼1.2−1.6 V	∼3.2−3.8 V
energy density (Wh kg^{-1})	50−135 (lab); ∼35−60 (practical stack-level)	∼160−250 (cell-level)
cycle life (cycles)	1000−6000 (lab); ∼6500 (DOE projection)	3000−8000 (depending on chemistry and depth of discharge)
round-trip efficiency (%)	∼70−80%	∼85−95%
levelized cost of storage (LCOS)	∼\$0.15−0.25 kWh^{-1} (baseline); target: <\$0.05 kWh^{-1}	∼\$0.13−0.18 kWh^{-1} (current utility-scale systems)
safety	nonflammable electrolytes; minimal thermal runaway risk	flammable electrolytes; thermal management required
manufacturing readiness	TRL: 5−7; MRL: 4−5	TRL: 9; MRL: 9 (mass production established)

[a] Abbreviations: VO_x: Vanadium oxides; NMC: $Li(Ni_xMn_yCo_z)O_2$; LFP: $LiFePO_4$; NCA: $Li(Ni_xCo_yAl_z)O_2$; LCO: $LiCoO_2$; EC: Ethylene carbonate; DMC: Dimethyl carbonate; TRL: Technology readiness level; MRL: Manufacturing readiness level.

Table 2. Zn Battery Cost and Performance (2030 Estimates)[a,14]

category	details
target LCOS	\$0.05 kWh^{-1}
2030 baseline LCOS	\$0.15 kWh^{-1} (excluding energy cost)
2030 baseline cycle life	6508 cycles
2030 baseline calendar life	17 years
2030 baseline round-trip efficiency	74%
key technical bottlenecks	energy density; low cycle life; moderate RTE; dendrite growth; gas evolution; limited voltage stability
high-impact innovation areas	Zn anode design, cathode optimization,
cycle life improvement	>400% (via cathode optimization); >240% (via Zn metal improvement)
round-trip efficiency improvement	∼30% (via Zn metal improvement); ∼27% (via electrolyte innovation)

[a] Abbreviations: LCOS: levelized cost of storage; RTE: round-trip efficiency.

200%.[14] Critically, many laboratory-scale innovations remain untested under commercial conditions such as lean electrolyte environments, high current densities, and pouch cell configurations. As such, this review builds a framework around two central metrics, energy density and cycle life, to evaluate the commercial relevance of AZIB materials research. Capacity is mainly determined by the cathode composition and reaction mechanisms, whereas cycle life is controlled by the stability of electrode−electrolyte interfaces, particularly solid electrolyte interface (SEI).[17,18] This review emphasizes the need for integrated and commercialization-aware strategies for electrolyte formulation, interface engineering, and component compatibility.[19−22]

This review presents the current state of AZIB research from a commercialization perspective, which aligns with DOE performance benchmarks such as energy density, cycle life, and cost.[9,10,23] The content mainly includes Mn-based and V-based cathodes, high-voltage electrolyte formulations, and zinc anode engineering strategies aimed at enhancing the electrochemical performance.[24−28] While zinc offers a high theoretical capacity (820 mAh g^{-1}) and intrinsic safety, issues such as hydrogen evolution and dendrite growth shorten the lifespan.[11,22,29] Similarly, cathode dissolution, sluggish ion diffusion, and structural instability are the barriers to achieving high energy density and cycle stability.[25,30] Aqueous electrolytes offer high ionic conductivity but suffer from narrow electrochemical stability window (ESW) and interfacial degradation.[8,31] Emerging strategies, such as additive incorporation, interphase engineering, and gel hybrid designs, are evaluated based on scalability, cost, and regulatory feasibility.[31−33] In addition, structural battery designs that integrate energy storage into mechanical components are a promising direction for wearable or multifunctional applications, but with trade-offs in mechanical durability and interface control. Collectively, as illustrated in Figure 1, this review offers a roadmap for transitioning AZIBs from lab-scale prototypes to commercially viable systems.

2. COMMERCIALIZATION BOTTLENECK: ENHANCING THE ENERGY DENSITY OF AZIBS

2.1. Cathode Capacity. As the demand for cost-effective and scalable energy storage technologies continues to grow, the commercialization of AZIBs hinges heavily on achieving competitive performance metrics, particularly in terms of energy density.[7,34] Among the key parameters, energy density is a crucial determinant of whether AZIBs can serve as viable alternatives to existing lithium- and sodium-based systems for large-scale utilization.[13,35] In this context, the development of high-performance cathode materials plays a pivotal role in enhancing the overall energy density of AZIBs.

MnO_2 has been widely used as a cathode material due to its high theoretical capacity, low cost, and environmental sustainability.[26,36] Nonetheless, its practical applications are limited by structural degradation, active material dissolution, and sluggish Zn^{2+} diffusion kinetics.[36−38] Strategies such as oxygen vacancy engineering, hybrid interface modifications, and electrolyte optimization have been adopted to mitigate these challenges.[26,39,40] However, the issues of poor scalability and long-term cycle stability of MnO_2 cathodes remain.[36,41,42] Similarly, vanadium-based cathodes offer high capacity and versatile redox behavior but face challenges such as dissolution in aqueous electrolytes, low operational voltages, and sluggish Zn^{2+} diffusion kinetics.[30,43] Defect engineering strategies, including the introduction of oxygen vacancies, cation doping, and structural modification, have shown great promise in enhancing ionic transport and structural stability.[28,44,45]

This section discusses cathode-related challenges in the commercialization of AZIBs and highlights advanced materials design and interfacial engineering strategies to enhance electrochemical performance. To establish a practical performance benchmark for cathode materials, we adopt an expected specific capacity range of 300−400 mAh g^{-1}. This estimation is grounded in system-level energy density targets, which call for energy densities above 150 Wh kg^{-1} for long-duration energy storage applications.[14] The energy density value is based on an average working voltage of approximately 1.45 V for Mn-based

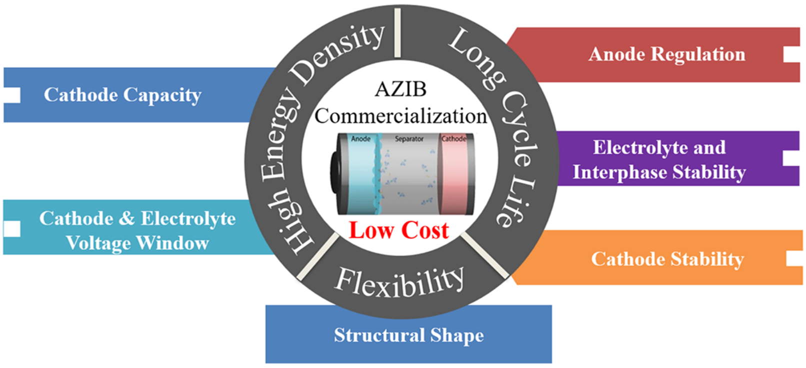

Figure 1. Schematic illustrating the progress of aqueous zinc-ion batteries toward commercialization.

Table 3. Electrochemical Performance of MnO_2-Based AZIBs[a]

material	voltage range (V)	electrolytes	capacity (mAh g^{-1})	cycling performance	RC/Rate	refs
Ile-α-MnO_2	0.8−1.8	2 M $ZnSO_4$ + 0.2 M $MnSO_4$	334.8 (0.1 A g^{-1})	85% after 2000 cycles at 1 A g^{-1}	167.1 at 1 A g^{-1}	37
Ca-Ile-α-MnO_2	0.4−1.9	3 M $Zn(CF_3SO_3)_2$ + 0.2 M $Mn(CF_3SO_3)_2$	289 (300 mA g^{-1})	80% after 1000 cycles at 1.5 A g^{-1}	50.6/5 A g^{-1}	38
PANI-MnO_2	0.9−1.8	2 M $ZnSO_4$ + 0.2 M $MnSO_4$ + Dioctyl Phthalate	∼2.5 Ah (pouch cell)	∼1-month reversible cycles at 7.5 mA cm^{-2}	0.5 Ah/7.5 mA cm^{-2}	53
MnO_2@N	1.0−1.8	2 M $ZnSO_4$ + 0.5 M $MnSO_4$	157 (0.5 A g^{-1})	100% after 500 cycles at 0.5 A g^{-1}	175/0.5 A g^{-1}	54
				83% after 1000 cycles at 5 A g^{-1}	124/5 A g^{-1}	
γ-MnO_2	1.0−1.8	2 M ZnSO4 + 0.1 M $MnSO_4$	493 (1 A g^{-1})	287 cycles with a peak capacity of 493 mAh g^{-1}	531/0.2 A g^{-1}	52
MnO_2@ZnMn-PBA	0.9−1.9	5 M $ZnSO_4$+ 0.6 M $MnSO_4$	370 (0.5 A g^{-1})	83% after 500 cycles at 1 A g^{-1}	372.6/0.5 A g^{-1}	39
				52.3 mAh g^{-1} after 50,000 cycles at 20 A g^{-1}	47.6/40 A g^{-1}	
K-MnO_2−NR	0.8−1.8	2 M $ZnSO_4$+ 0.1 M $MnSO_4$	285 (0.1 A g^{-1})	222 mAh g^{-1} at 2 A g^{-1}	285/0.1 A g^{-1}	51
				Negligible decay after 1450 cycles at 2 A g^{-1}	222/2 A g^{-1}	
DOE target			300−400	≥6500 (LDES)		14

[a] Abbreviation: LDES: long-duration energy storage.

cathodes and assuming that cathode active materials account for roughly 20−40% of the total cell mass.[24−26] By analyzing current research findings and proposing future research directions, this section aims to provide insights into developing robust, high-performance AZIBs for large-scale energy storage applications.

MnO_2 exhibits various polymorphic structures, including α-, β-, γ-, and δ-phases, enhancing its versatility for electrochemical energy storage applications.[46,47] Nonetheless, the energy density of MnO_2 cathodes remains constrained by incomplete utilization of redox reactions and limited voltage plateaus, which hinder their deployment in large-scale electrochemical energy storage systems.[24−26] A major obstacle is the insufficient structural robustness required to sustain deep discharge reactions needed to fully extract the theoretical capacity.[26,36] To address these issues, many engineering strategies have been adopted, including oxygen vacancy engineering, hybrid interface modifications, and the incorporation of stabilizing additives into electrolytes[25,48−51] (Table 3). To increase the specific capacity of MnO_2-based cathodes in AZIBs, Li et al.[52] introduced a composite material (MNC-50) by growing δ-MnO_2 nanoflowers on residual carbon (RC). The RC was extracted from coal gasification slag via a simple acid leaching-hydrothermal process (Figure 2a). Benefiting from enhanced conductivity, abundant oxygen vacancies, and a highly open nanostructure, the MNC-50 cathode delivered a capacity of 432 mAh g^{-1} at 0.1 A g^{-1}, outperforming pristine δ-MnO_2 (Figure 2b). As illustrated in Figure 2c, the RC framework effectively supports Zn^{2+}/H^+ cointercalation and structural stabilization during cycling, offering a scalable, waste-derived strategy for constructing high-capacity and durable cathodes in AZIBs. While this capacity exceeds the expected benchmark of 300−400 mAh g^{-1}, its rate performance and long-term cycle stability under practical loadings remain essential for full alignment with energy density targets.[14]

Vanadium-based cathodes are attractive for AZIBs due to their high theoretical capacity (∼400 mAh g^{-1}), rich redox chemistry, and structural adaptability.[28,30] Table 4 shows the summary of the electrochemical performance of typical reported composite vanadium-based cathodes for AZIBs.

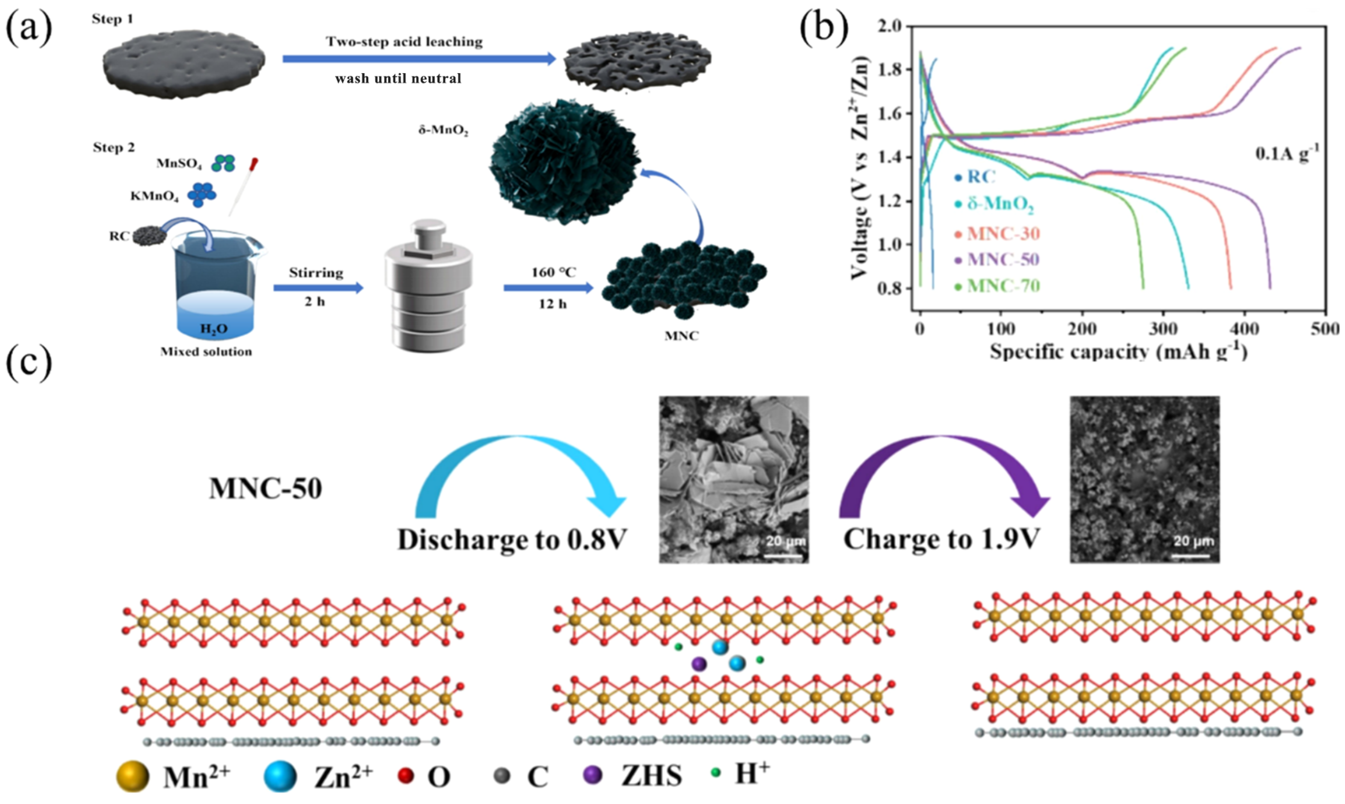

Figure 2. (a) Preparation process of δ-MnO_2/residual carbon composite and δ-MnO_2. (b) Charge/discharge profiles of δ-MnO_2, RC, MNC-30, MNC-50 and MNC-70. (c) Schematic diagram of ionic transport mechanism in MNC-50 cathode. Reproduced with permission from ref 52. Copyright 2024, Elsevier.

Table 4. Electrochemical Performance of Vanadium-Based AZIBs[a]

material	voltage range (V)	electrolytes	capacity (mAh g^{-1})	cycling performance	RC/Rate	refs
VO_2-D (Tunnel-Oriented)	0.1−1.6	3 M $Zn(CF_3SO_3)_2$	420.8 (0.1 A g^{-1})	84.3% after 5000 cycles at 10 A g^{-1}	344.8/10 A g^{-1}	61
PAVO-1	0.6−1.8	3 M $ZnSO_4$	350 (0.1 A g^{-1})	90% after 100 cycles at 0.1 A g^{-1}	190/5 A g^{-1}	45
Od-VO_2	0.2−1.4	3 M $ZnSO_4$	202.8 (0.5 A g^{-1})	90.1% after 1000 cycles at 1 A g^{-1}	143/5 A g^{-1}	62
E-VON	0.3−1.9	3 M $ZnSO_4$	305 (0.1 A g^{-1})	73% after 2500 cycles at 10 A g^{-1}	132 Wh kg^{-1} at 8500 W kg^{-1}	58
Ni_2−NH_4−V_2O_5	0.3−1.8	3 M $ZnSO_4$ + 0.1 M $NiSO_4$	320 (0.5 A g^{-1})	91% after 2000 cycles at 1 A g^{-1}	215/2 A g^{-1}	59

[a] Abbreviation: PAVO: PANI-intercalated V_2O_5; E-VON: electrically activated NH_4^+-intercalated vanadium oxide.

However, commercialization is limited by challenges such as structural degradation, vanadium dissolution, sluggish Zn^{2+} diffusion, and low working voltage (~0.8 V vs Zn/Zn^{2+}).[43,55,56] Water-induced side reactions and poor structural resilience often lead to vanadium loss and capacity fading.[57] Additionally, the strong electrostatic interaction between hydrated Zn^{2+} and vanadium-based hosts hinders ion transport and destabilizes the lattice.[43,56] Recent advances in defect engineering have shown promise in mitigating these issues. Strategies such as introducing oxygen vacancies, doping (e.g., N, NH_4^+, Na^+), and water intercalation expand interlayer spacing, enhance Zn^{2+} diffusion and reduce dissolution of vanadium-based cathode.[44,45,58,59] Lattice amorphization has also been shown to generate new ionic diffusion pathways and improve rate performance.[60] While these approaches improve both energy density and structural stability, the lack of scalable and cost-effective synthesis methods remains a major obstacle to widespread implementation.[27,30,57] Although some vanadium-based cathodes have demonstrated specific capacities exceeding 350 mAh g^{-1} in lab-scale tests, their voltage plateau (~0.8−1.2 V) and full-cell energy densities still fall short of the >150 Wh kg^{-1} for long-duration storage applications. The future research aims to integrate these strategies with a focus on interfacial dynamics and long-term cycle stability, paving the way for the practical application of vanadium-based cathodes in next-generation AZIBs.

2.2. Enlarging the Electrochemical Stability Window of Electrolytes. A critical barrier to enhancing the energy density of AZIBs is the narrow ESW of conventional water-based electrolytes that are typically limited to ~1.23 V by the onset of hydrogen and oxygen evolution.[8,20,63] The narrow ESW restricts the operating voltage of AZIBs and limits the selection of compatible high-voltage cathodes, resulting in inferior energy densities compared to commercial lithium

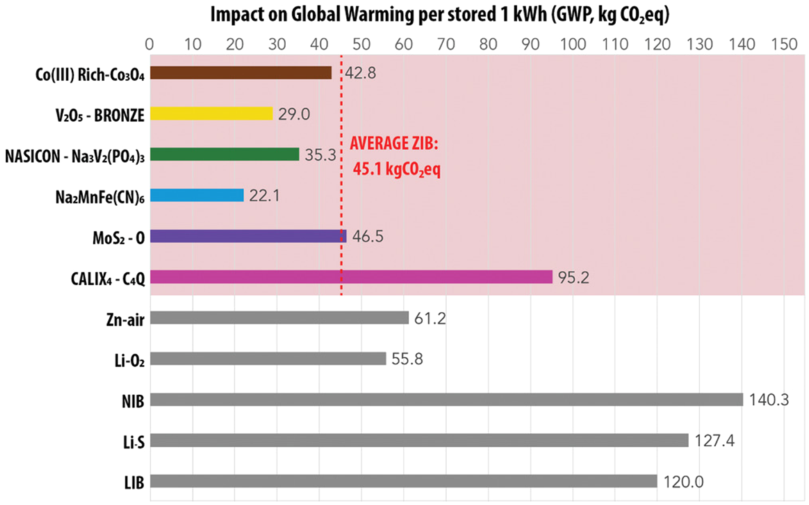

Figure 3. Global warming of six aqueous zinc ion batteries standardized to 1 kWh of storage capacity. For the sake of comparison, data corresponding to Zn-air batteries is taken from reference 71; data corresponding to Li−O_2 batteries is taken from reference 72; data corresponding to NIBs is taken from reference 73; data corresponding to Li−S batteries is taken from reference 74; data corresponding to LIBs is taken from reference 75. Reproduced with permission from ref 69. Copyright 2021, Wiley.

batteries.[10,33,63] Recent efforts in laboratory research have focused on expanding the ESW through electrolyte engineering, such as developing water-in-salt electrolyte (WiSE) systems, hybrid organic-aqueous solvents, and deep eutectic formulations, enabling suppressed water decomposition and higher-voltage operation.[64,65] As the ESW fundamentally defines the upper voltage limit of an AZIB cell, its optimization plays a pivotal role in maximizing energy output.[66] Therefore, extending the ESW is not only a key electrochemical challenge but also a crucial step for meeting commercial performance targets in high-energy storage applications.

Widening the ESW is not only a scientific pursuit but a commercial imperative for AZIBs. The narrow ESW of conventional aqueous electrolytes fundamentally limits the cell voltage and energy density (>150 Wh kg^{-1}). Although strategies like water-in-salt, deep eutectic systems, and hybrid aqueous−organic electrolytes have extended ESWs to ∼2.5−3.0 V in lab-scale demonstrations, most of these systems suffer from high viscosity, cost, or poor compatibility with scalable manufacturing.[65,67] Moreover, many reported high-voltage systems require excess electrolyte or customized electrodes that deviate from industry standards. For commercialization, electrolyte systems must maintain a widened ESW while meeting critical constraints such as low cost, environmental safety, compatibility with lean electrolyte usage, and stability under pouch cell operation.[64,65,68] Bridging this gap requires electrolyte formulations that integrate interfacial stabilizers, suppress water decomposition, and remain functional under high areal capacities and industrial cell formats. As such, the ESW must be optimized not only for voltage expansion but also for integration feasibility, ensuring AZIBs can meet long-duration storage and grid deployment requirements.[64,65,67]

2.3. Environmental Sustainability and Life Cycle Considerations of AZIBs. AZIBs are increasingly recognized for their potential environmental advantages over conventional LIBs, primarily due to their use of abundant materials and safer aqueous electrolytes. A cradle-to-gate life cycle assessment (LCA) of six laboratory-scale AZIB designs revealed an average global warming potential (GWP) of 45.1 kg CO_2-equivalent per kWh, which is competitive with or superior to that of LIBs. This lower GWP is attributed to the elimination of flammable organic solvents and the avoidance of energy-intensive dry-room assembly processes.[69,70]

However, certain components, notably the cathode and separator, contribute significantly to the overall environmental impact (Figure 3). The cathode's multistage fabrication process and the separator's requirement for mechanical robustness, often achieved through energy-intensive materials like glass microfiber, are key factors. To mitigate these impacts, research is focusing on developing cathodes that operate at higher voltages with increased mass loadings, thereby reducing material usage and improving energy density.[76] Additionally, the exploration of renewable materials, such as polysaccharide-based hydrogels, for electrolytes and separators offers promising avenues for enhancing the sustainability of AZIBs.[69,76]

Recycling is another critical aspect of AZIB sustainability. While still in its early stages, studies have demonstrated that metallic zinc can be recovered with over 99% efficiency through evaporation and separation processes, allowing for indefinite recycling without degradation of physical properties. The development of biopolymer electrolytes further facilitates environmentally friendly recycling approaches, as these materials can be processed using benign methods.[77] Moreover, the use of organic electroactive materials that are soluble in inexpensive organic solvents and stable in various charge states enables high recovery potential at any state of charge. In conclusion, while AZIBs present a promising path toward sustainable energy storage, ongoing efforts to optimize component materials, manufacturing processes, and end-of-life recycling are essential to fully realize their environmental benefits.[69,71,77]

3. COMMERCIALIZATION BOTTLENECK: ENHANCING THE CYCLE LIFE OF AZIBS

To enable the widespread commercialization of AZIBs, improving long-term cycle stability is crucial.[20,66] According to the U.S. DOE's LDES goals, systems must demonstrate stable operation over thousands of cycles with minimal capacity fade to meet LCOS targets below \$0.05 kWh^{-1}.[14]

Table 5. Electrochemical Performance of AZIBs with Different Electrolyte Engineering Strategies

strategy/method	electrolyte/additive	initial capacity (mAh/g or mAh)	cycling stability (retention, rate)	refs
ultrahigh-modulus hydrogel electrolyte	PVA/PAM/PAN hydrogel + 2 M $ZnSO_4$ (~70% H_2O)	~230@0.2 A g^{-1}	1000 cycles, ~90% retention@1A g^{-1}	98
trifunctional SADS additive	2 M $ZnSO_4$ + 0.2 M SADS	253@1 A g^{-1}	1000 cycles, 80%retention@1 A g^{-1}	99
cationic surfactant (DTAC) interfacial mod.	2 M $ZnSO_4$ + 10 mM DTAC	149@5 A g^{-1}	2000 cycles, 83%retention@5 A g^{-1}	100
low-polarity cosolvent modulation	3 M $Zn(ClO_4)_2$ in DGM/H_2O (6:4)	88.8@1 A g^{-1} (−20 °C)	10,000 cycles, ~100% retention@ 1 A g^{-1} (−20 °C)	101
wide-temp/mechanical-adaptive hydrogel	2 M $ZnSO_4$ in H_2O/DMF (20% DMF) + PAM hydrogel	135@1 A g^{-1}	3000 cycles, 81.7% retention@1 A g^{-1} (25 °C) 750 cycles, 92.5%@0.1 A g^{-1} (−20 °C)	102
HDES-induced gradient SEI (wide temp)	$Zn(OTf)_2$ + ethylene glycol + H_2O + SnI_4 (HDES)	237@1 A g^{-1}	10,000 cycles, 79% retention@10 g^{-1}	103
polymer−inorganic bilayer SEI (proton holder)	2 M $Zn(OTf)_2$ + aniline additive	1.2 Ah (initial, pouch cell)	350 h (~78% retention), 335 mAh/507 h@30°C/72% retention	104
ABPA-induced multifunctional SEI	1 M $Zn(OTf)_2$ + 5 mM ABPA	~49 mAh (pouch cell)	80.3% retention after 300 cycles, 61.2% after 1000 cycles	105
fluorine-rich SEI	2 M $Zn(OTf)_2$ + 5% 1,4-thioxane	6.8 mAh cm^{-2} (areal)	98% retention after 400 cycles (high mass loading, lean E/C)	17

In AZIBs, while zinc metal anodes and aqueous electrolytes provide inherent cost and safety benefits, their practical implementation is hindered by severe degradation during repeated cycling.[11,42,63] These issues include hydrogen evolution reaction, zinc dendrite formation, electrolyte decomposition, and irreversible cathode dissolution.[19,66,78,79] The issues lead to capacity fading, decreased Coulombic efficiency, and safety concerns, ultimately limiting the operational lifespan of the battery and its commercial viability.[12,80]

Among these challenges, interfacial instability between the electrode and electrolyte has emerged as a dominant factor affecting the cycle life of AZIBs.[53,81] Repeated Zn^{2+} plating/stripping, along with dynamic electrolyte interactions, can result in side reactions, passivation layer formation, and gas evolution. These processes degrade the SEI on the anode and cathode-electrolyte interphase (CEI) on the cathode, impairing ionic transport and electrode integrity.[17,18,82] Addressing these issues requires an integrated strategy involving anode surface engineering, electrolyte design, and controlled SEI/CEI formation.[17,83] The following sections explore each of these perspectives in detail, outlining recent progress and ongoing challenges in stabilizing the electrode−electrolyte interface to improve cycle life and support the commercial transition of AZIB technology, as shown in Table 5.

Waste battery recycling is also essential for commercialization. Since mass production for commercialization requires a large amount of metal resources, environmental pollution during metal mining is inevitably severe. In particular, transition metals (e.g., Mn, V, Co, Ni) of main constituents in electrodes are highly related to environmental implications as well as economic issues.[69] Pyrometallurgy, hydrometallurgy and direct recycling are broadly utilized for metal recycling, and these approaches enable the recycling of the waste battery to reduce carbon emission dramatically compared to primary mining. In addition, recycling of these transition metals can support supply chain resilience by replacing dependency on primary raw materials with recycled sources.[69]

3.1. Electrolytes and Interfacial Engineering.

AZIBs face considerable commercialization challenges primarily due to limited electrolyte stability, particularly issues associated with parasitic side reactions and the formation of unstable interphases at the electrode−electrolyte interface.[81] The performance and longevity of aqueous electrochemical systems depend critically on electrolyte stability since electrolyte decomposition can trigger adverse reactions and promote uncontrolled zinc dendrite growth on the anode surface. Despite the high ionic conductivity and intrinsic safety, aqueous electrolytes are susceptible to side reactions due to the limited ESW of water. When AZIBs operate near or beyond the ESW, water decomposition can initiate parasitic reactions, such as hydrogen evolution, pH fluctuation, and the formation of insulating byproducts.[84−86] These reactions compromise the chemical stability of the electrolyte and, more importantly, hinder the formation and maintenance of a stable SEI on the zinc anode surface. An unstable SEI cannot effectively regulate ion transport or protect the anode from ongoing corrosion, resulting in increased internal resistance, reduced Coulombic efficiency, and accelerated capacity degradation.[84,87,88]

Another electrolyte-related challenge impeding the commercialization of AZIBs is limited ion transport, particularly under a high current density. Efficient and sustained ion mobility is essential for applications requiring rapid charge−discharge rates and high-power output, such as electric vehicles and grid-scale energy storage systems.[89,90] At elevated current densities, the efficiency of ion mobility within the electrolyte decreases due to increased viscosity and potential concentration gradients. These issues lead to reduced power output and diminished cyclic stability, which is detrimental to the overall operational lifespan of the battery.[84,91] Addressing these transport limitations requires innovative electrolyte formulations that enhance ionic conductivity while reducing internal resistance.[92] The development of advanced polymers or ionic liquids as electrolytes shows promise to overcome these bottlenecks, thereby enabling AZIBs to meet the growing demand for high-performance energy storage devices.[93−96]

To overcome the challenges facing electrolyte stability, research has increasingly focused on the development of advanced electrolyte formulations and additives that suppress side reactions at the electrode−electrolyte interface and further enhance the overall electrochemical stability of AZIBs.[84,87,88] Swelling clays such as laponite (Figure 4a) have emerged as

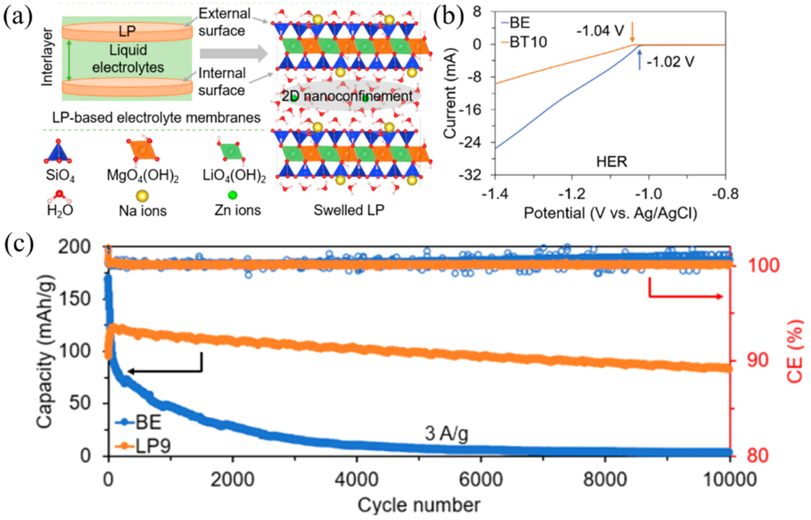

Figure 4. (a) Schematic illustrations of electrolyte membranes based on LP nanocrystals with 2D nanostructures and exceptional swelling capabilities. (b) Linear sweep voltammetry curves of baseline and BT10 electrolytes. Reproduced with permission from ref 85. Copyright 2023, Wiley. (c) Long-term cycling performance at 3 A g^{-1}. Reproduced with permission from ref 86. Copyright 2023, American Chemical Society.

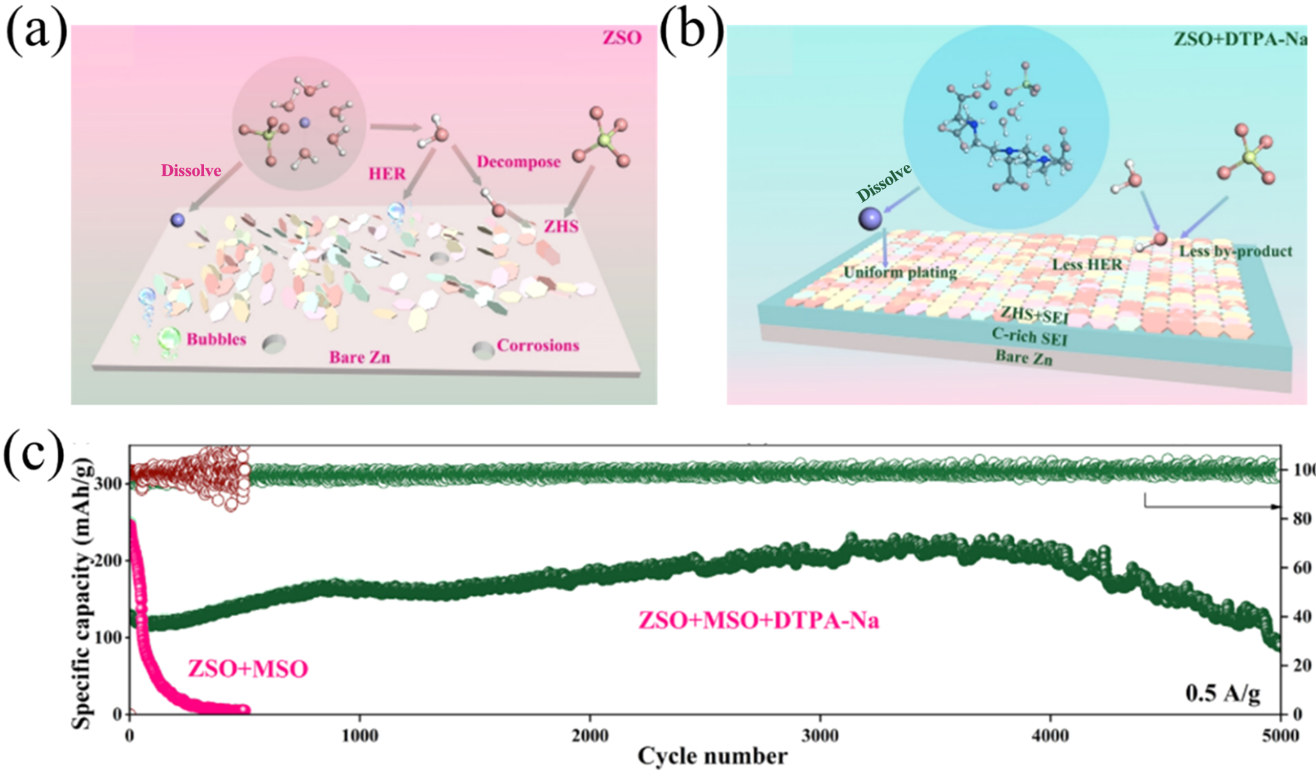

Figure 5. (a, b) Schematic illustration of the underlying mechanism of enhanced electrochemical performance by the addition of diethylenetriamine pentaacetate sodium (DTPA-Na). (c) Long-term cycling performance at 0.5 A g^{-1}. Reproduced with permission from ref 82. Copyright 2024, Wiley.

promising frameworks for stabilizing aqueous electrolytes in AZIBs by reducing water activity and suppressing interfacial side reactions.[86] Laponite-based "water-in-swelling-clay" electrolytes (WiSCE) demonstrated efficacy in suppressing hydrogen evolution, zinc corrosion, and the formation of zinc hydroxysulfate (ZHS) byproducts (Figure 4b). As a result, laponite-supported quasi-solid-state systems have demonstrated exceptional cycle stability, achieving over 10,000 cycles with minimal capacity loss at high current densities, as shown in Figure 4c. Moreover, the low cost and natural abundance of swelling clays like bentonite-based WiSCE system[85] offer a striking cost advantage. The estimated material cost is only \$924 per ton, which is over 7000 times cheaper than state-of-the-art WiSCE electrolytes. The low price highlights its strong potential for scalable deployment in grid-scale energy storage.

Meanwhile, multifunctional additives have been introduced to enhance zinc-ion solvation and regulate dendrite growth.[84,87] However, scaling innovative solutions for commercial production faces hurdles, such as ensuring consistent manufacturing processes while maintaining cost-

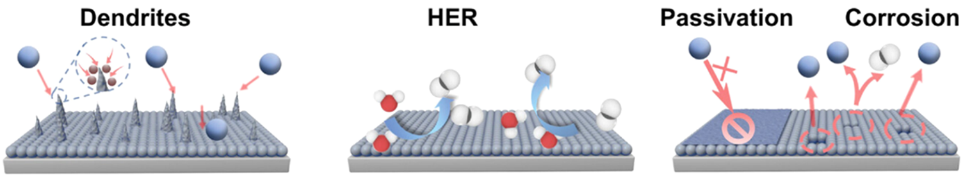

Figure 6. Schematic diagram of parasitic reactions including dendritic growth, hydrogen evolution reaction, and water-induced corrosion reaction. Reproduced with permission from ref 106. Copyright 2024, Wiley.

effectiveness. For instance, diethylenetriamine pentaacetate sodium (DTPA-Na) was introduced as a multifunctional electrolyte additive capable of simultaneously regulating the Zn^{2+} solvation structure and constructing synergistic interfacial layers on both electrodes.[82] As illustrated in Figure 5a, this strategy facilitates the formation of a dense, multilayered SEI that suppresses hydrogen evolution and dendrite growth, while Figure 5b reveals the altered solvation structure that underpins this interfacial stability. More importantly, a CEI forms in situ on the MnOOH surface, effectively inhibiting Mn dissolution and enabling a full-cell lifespan exceeding 5000 cycles at 0.5 A g^{-1}, as shown in Figure 5c. These results demonstrate that interface-wide additive engineering provides a practical route to durable and scalable AZIB systems by enhancing both electrode stability and cycle longevity. Consequently, addressing the challenges associated with electrolyte stability is pivotal for the successful commercialization of AZIBs, necessitating collaborative research and development efforts to unlock their full potential in energy storage applications.[20,97]

The incorporation of functional electrolyte additives has proven highly effective in enhancing the electrochemical performance of AZIBs, particularly by improving ionic conductivity, stabilizing electrode interfaces, and mitigating unwanted side reactions. Despite these advantages, the integration into electrolytes presents challenges in terms of cost, scalability, and regulatory compliance. Most electrolyte additives, especially those based on complex organic or rare elements, significantly increase overall battery manufacturing costs. To enable the widespread adoption of AZIBs, these cost concerns must be addressed through comprehensive consideration of raw material sourcing, large-scale production feasibility, and potential cost-reduction strategies, such as industry partnerships or recycling initiatives. For instance, many high-performance additives, particularly organic or fluorinated compounds, are priced above \$100 kg^{-1} in laboratory settings, far exceeding the target of \$10 kg^{-1} cost target for practical grid-scale batteries.[8,19,20,32,84]

Beyond cost considerations, regulatory barriers are significant obstacles that must be addressed when adopting electrolyte additives for commercial AZIB deployment. Many additives, particularly those involving volatile organic compounds or potentially toxic elements, face stringent environmental regulations that limit their applicability in commercial settings.[8,32,33] Compliance with safety standards and environmental guidelines requires significant investment in testing and certification processes that further increase production costs and delay commercialization. These regulatory challenges necessitate the careful selection of additives that are not only effective in enhancing performance but also safe, environmentally benign, and compliant with current industry standards.[31,32]

3.2. Anode Regulation.

Zinc metal anodes are fundamental to the design and performance of AZIBs, yet their commercial viability is hindered by critical issues such as dendrite formation, hydrogen evolution reaction (HER), and surface corrosion, as shown in Figure 6.[11,29,106] Among these, zinc dendrites form from uneven zinc deposition during plating/stripping cycles, potentially causing internal short circuits, electrolyte leakage, and even catastrophic cell failure.[80,107] These risks become more acute under high current densities or prolonged cycling, conditions commonly encountered in commercial applications such as grid-scale energy storage and electric mobility. To suppress dendrite formation, various surface modification strategies have been developed. Techniques such as laser-induced fluorine-containing carbon coatings[108] have demonstrated strong potential for stabilizing Zn anodes in AZIBs by constructing robust interphases with low resistance and high cycling durability. However, the practical implementation of these methods in commercial-scale production is not ideal. The challenges mainly include the scalability of fabrication techniques, the chemical compatibility of interface materials with aqueous electrolytes, and the complexity introduced into manufacturing workflows.

In parallel, HER and corrosion further reduce Coulombic efficiency and accelerate electrode degradation.[85,86] Parasitic HER arises from the interaction between water molecules and zinc metal, leading to unwanted hydrogen gas accumulation, increased internal pressure, and thus structural instability.[80,109] It also induces localized pH fluctuations that exacerbate zinc corrosion, further compromising the long-term stability of the anode.[107,110,111] To mitigate these effects, researchers have adopted various electrolyte additives capable of forming protective interfacial layers that suppress HER and enhance zinc deposition uniformity.[32,88,92,95] While these strategies have exhibited enhanced battery lifespan and performance at the laboratory scale, their long-term effectiveness in industrial-level applications is uncertain. Corrosion-induced passivation of the zinc anode significantly compromises battery stability and manufacturing consistency. The formation of passivation layers obstructs ion transfer, reduces cycling performance, and accelerates capacity fading. Furthermore, in corrosive environments, particularly under high-voltage operations, the accumulation of byproducts (e.g., ZHS) can further block ion pathways and degrade electrochemical performance. Overcoming these challenges requires an integrated approach that combines material innovation, advanced manufacturing techniques, and strategic electrolyte engineering. Addressing these issues is crucial to achieving the performance, safety, and cost targets necessary for the widespread commercialization of AZIBs.

However, beyond interfacial stability, the most critical and under-addressed issues for Zn anode commercialization lie in achieving sufficient areal capacity and a high Zn utilization ratio (ZUR).[107,110] Although high current densities are often adopted in lab-scale tests to demonstrate long-term cycling,

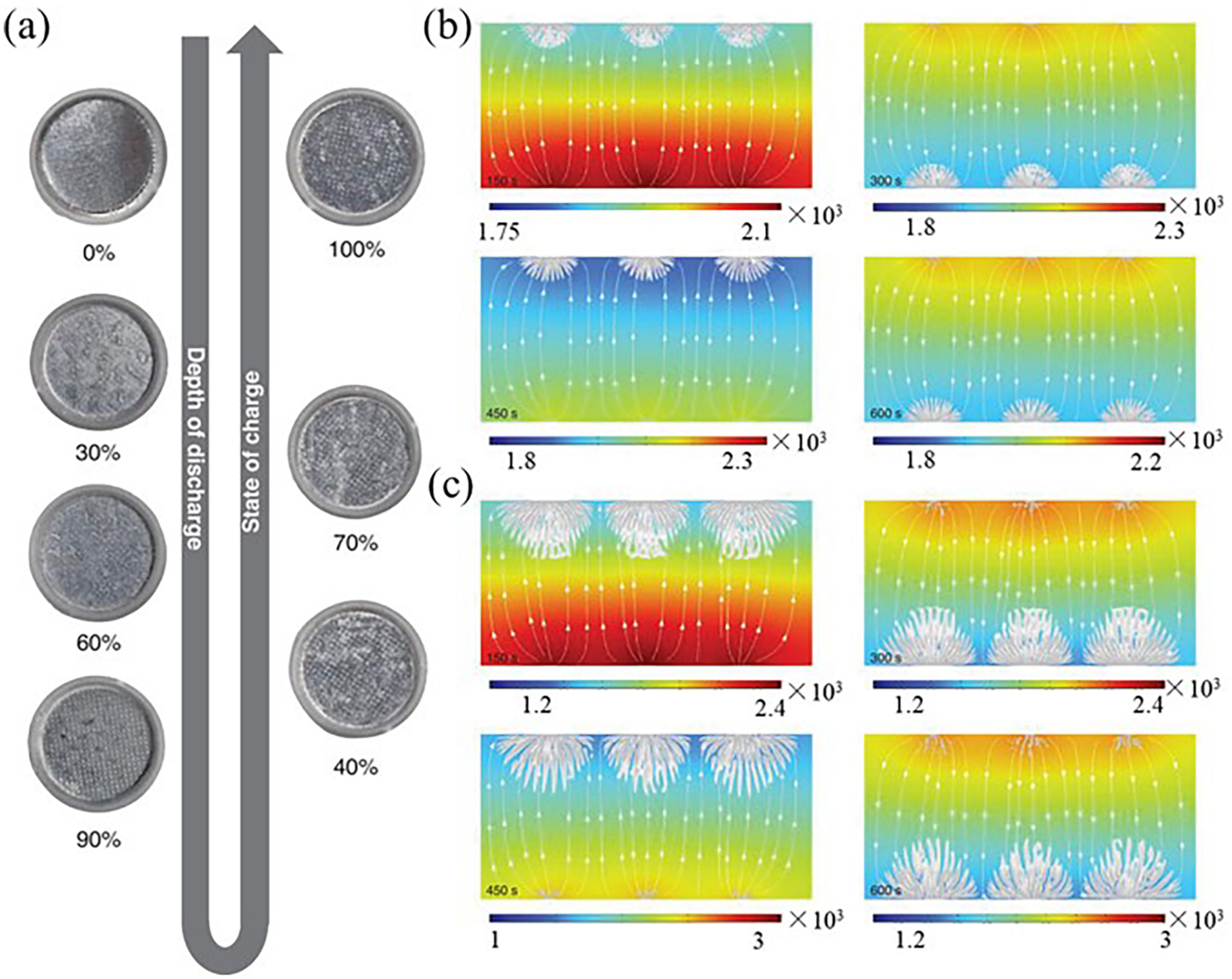

Figure 7. (a) Morphological changes of Zn anode undergoing continuous discharge (different depth of discharge, DOD) and subsequent charging processes (different states of charge, SOC). Where DOD = 100% − SOC. COMSOL simulation of Zn metal plating and stripping processes at a constant current density of (b) 10 mA cm^{-2} and (c) 40 mA cm^{-2}. The color variation represents the concentration change of Zn^{2+}. Reproduced with permission from ref 12. Copyright 2023, Wiley.

these conditions typically correspond to unrealistically low areal capacities (<1 mAh cm^{-2})[23] and mask the true instability of Zn metal under more practical loadings. While higher current densities may improve Zn nucleation density and reduce dendrite formation due to elevated overpotentials, these benefits often do not translate under high-capacity or thick-Zn configurations. Moreover, the widespread use of thick Zn foils (>50 μm)[112] leads to low depth of discharge (DOD), often <5%, which severely limits the energy density of full cells. Xu et al.[12] addressed the challenge of Zn anode degradation under high ZUR by identifying structural collapse as the primary failure mechanism, which was demonstrated by severe Zn depletion and irreversible morphology collapse (Figure 7a). Simulations and experimental results show that lower current densities (e.g., 10 mA cm^{-2}) maintain structural integrity (Figure 7b), while higher densities (e.g., 40 mA cm^{-2}) induce dendritic growth and significant electrode thickening (Figure 7c). These findings emphasize the importance of mechanical stability in evaluating Zn anodes under practical ZUR conditions.

Balancing high areal capacity with acceptable Zn utilization is critical but rarely achieved in current research. When Zn is deeply cycled with high DOD, severe morphological damage such as partial delamination and uneven redeposition occurs, as evidenced by structural failure under high-DOD conditions.[113] Conversely, low-DOD systems avoid such damage but offer limited insight into full-cell relevance or energy output. Hence, performance assessments of Zn anodes under either extremely thick or ultrathin configurations may be misleading. As a result, rational design may need to consider both capacity per area and Zn thickness/DOD simultaneously. Systematic studies conducted at moderate current densities with realistic cathode/anode loadings are essential to properly evaluate interfacial stability, failure mechanisms, and the commercial potential of Zn metal anodes in AZIBs.[11,113]

3.3. Cathode Stability.

Despite the widespread investigation of MnO_2 as a cathode material for AZIBs, its practical application is constrained by intrinsic challenges. Specifically, the dissolution-redeposition behavior of Mn species, often observed during cycling, contributes to poor long-term stability and material loss.[26,39,114] Furthermore, the inherent complexity

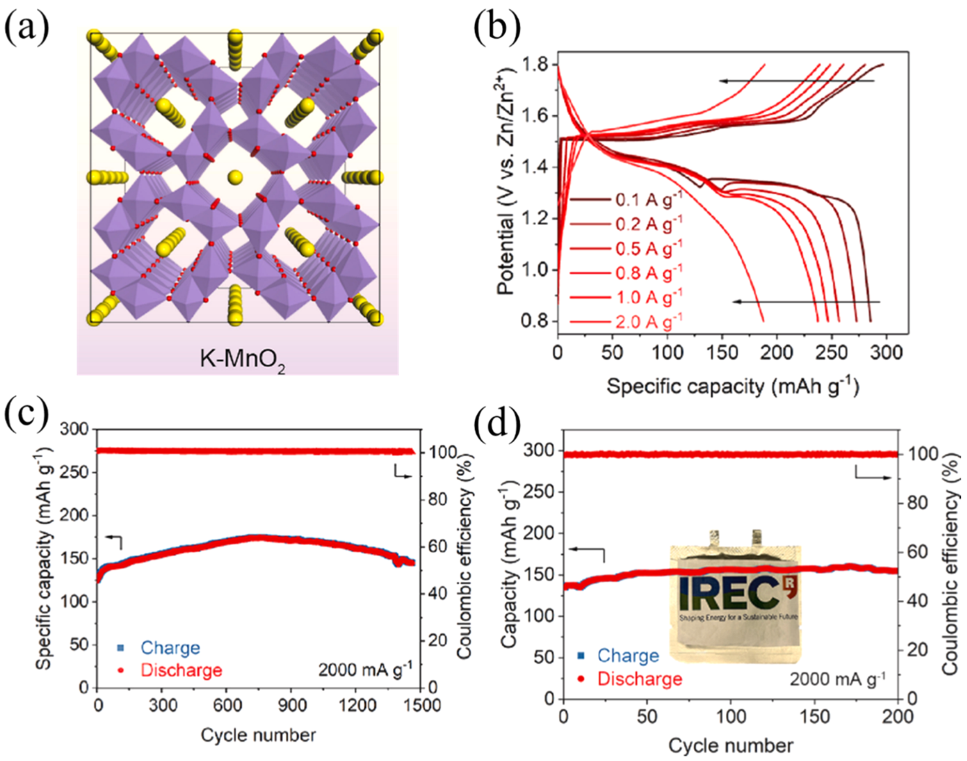

Figure 8. (a) Illustration of K-ion intercalation into α-MnO_2. (b) Galvanostatic charging/discharging (GCD) curves at various current densities. (c) Long-term cycling performance at 2 A g^{-1}. (d) Cycle stability of the pouch cell at 2 A g^{-1}. Reproduced with permission from ref 51. Copyright 2024, Elsevier.

of the MnO_2 energy storage mechanism continues to hinder its commercialization.[36,115,116] The theoretical model of Zn^{2+} and proton coinsertion, enabling access to its second-electron redox capacity, can favorably enhance the energy density of AZIBs.[49,117,118] However, it also leads to increased structural instability and poor reversibility during extended cycling.[119] This limitation necessitates innovative material designs and manufacturing processes to achieve both high practical capacity and long-term stability. Recent advancements, such as hybrid organic−inorganic interfaces, cation doping,[38,120] and composite cathode materials, have shown great promise in stabilizing multielectron reactions while maintaining the structural integrity of MnO_2. However, these modifications often introduce additional costs and synthesis complexities, posing significant challenges to cost-effective scalability in the commercialization process.

Moreover, an integrated approach that combines structural engineering, electrolyte optimization, and cathode/electrolyte interface modifications is necessary. For instance, the incorporation of carbon-based materials, such as graphene or conductive polymers, has been explored to enhance electronic conductivity and mitigate Mn dissolution from the cathode material. Additionally, preintercalation of cations into MnO_2 frameworks helps to stabilize the crystal structure, preventing phase collapse and thus improving cycle stability.[48,51,54] Chen et al.[51] developed a K^+ preintercalated nanorod architecture (K-MnO_2−NR) via a simple one-step redox process, as illustrated in Figure 8a. The resulting material features a well-aligned tunnel structure and ultrathin rod morphology, enabling short Zn^{2+} diffusion paths and high surface reactivity. As shown in Figure 8b, K-MnO_2−NR exhibits distinct and reversible voltage plateaus and achieves a high specific capacity of 222 mAh g^{-1} at 2 A g^{-1} with negligible decay over 1450 cycles (Figure 8c). Moreover, pouch cells with the cathode maintain stable performance at 2 A g^{-1} over 200 cycles (Figure 8d), demonstrating that the cathode is promising for scalable and cost-effective deployment in practical AZIB systems. This remarkable performance is attributed to the synergy between structural confinement, improved electronic pathways, and minimized volume changes during cycling. However, the scalability of current modification strategies, including oxygen vacancy engineering and organic−inorganic hybridization methods, is constrained by complex synthesis processes and high production costs.[40,48,121] As such, there is a continuing need to further develop novel cathode materials and interfacial engineering methods that are low-cost and highly scalable.

Apart from directly modifying cathode materials, electrolyte optimization has been explored to enhance the electrochemical performance of MnO_2 cathodes.[36,122,123] The introduction of Mn^{2+} to aqueous electrolytes helps mitigate dissolution by maintaining a dynamic equilibrium between MnO_2 and Mn^{2+}. However, this strategy still suffers from limited long-term effectiveness due to the gradual accumulation of inactive byproducts and side reactions that can disrupt the equilibrium over extended cycling.[5] Moreover, excessive Mn^{2+} concentrations can lead to unwanted precipitation reactions, forming Mn-based byproducts that reduce the availability of active Mn^{2+} species for continuous redox cycling.[26,39,46]

While substantial progress has been made in addressing the structural and electrochemical limitations of MnO_2, future research should prioritize cost-effective and scalable solutions to enable the practical deployment of the cathode material in grid-scale energy storage. Advancements in cathode material engineering and electrolyte design will be essential for overcoming these barriers and achieving the widespread adoption of MnO_2-based AZIBs. Achieving an optimal balance between structural stability, charge storage efficiency, and manufacturability remains a crucial challenge. Future efforts may benefit from the synergistic integration of multiple stabilization strategies, including defect engineering, surface modification, and tailored electrolyte formulations, to suppress active material dissolution and enhance interfacial stability for large-scale energy storage applications.[124]

3.4. Pouch Cell Performance. To bridge the gap between laboratory research and practical applications, it is imperative to evaluate AZIB performance under varying testing protocols, particularly comparing coin cells and pouch cells, as well as the impact of electrolyte quantity.[24,84,110,122] Coin cells, commonly used in academic studies, often employ excess electrolyte and low cathode loading, which can overstate performance metrics such as capacity retention and cycle life. In contrast, pouch cells, which more closely mimic commercial battery configurations, typically operate under lean electrolyte conditions and higher cathode loadings, presenting challenges such as increased internal resistance and reduced ion transport efficiency.[125−127] For instance, studies have shown that AZIBs tested in pouch cells with lean electrolyte exhibit faster capacity fading compared to their coin cell counterparts, highlighting the necessity for electrolyte optimization in practical settings.[68] Therefore, to accurately assess the commercial viability of AZIBs, it is essential to conduct performance evaluations under conditions that closely resemble real-world applications, including high mass loading electrodes and limited electrolyte volumes.[83,103]

While coin cells are the predominant format for evaluating AZIB performance in laboratory studies, their reported electrochemical metrics, such as specific capacity and cycle life must be critically interpreted due to highly favorable test conditions. Coin cells typically utilize an excess electrolyte amount, low cathode areal loading (<2 mg cm^{-2}), and an oversized zinc anode, resulting in minimal concentration polarization and negligible electrolyte depletion effects, thus significantly enhancing observed cycling stability and energy density metrics. In contrast, pouch cells, which closely emulate commercial battery designs, require higher electrode loadings (>10 mg cm^{-2}), limited electrolyte volume, and balanced N/P (negative-to-positive) ratios to achieve meaningful volumetric and gravimetric energy densities.[128] As reported in recent studies, such realistic conditions accelerate electrolyte depletion, interfacial deterioration, and electrode mechanical degradation, reducing cycle life from thousands of cycles typically demonstrated in coin cells to merely hundreds or fewer in pouch cells. For instance, even robust manganese oxide cathodes exhibiting ultralong cycle lives in coin-cell configurations (long cycles at high rates) have shown significantly diminished cycle stability (e.g., <1000 cycles) when integrated into pouch cells with commercially relevant conditions.[41,129] This disparity clearly indicates that scaling AZIBs from coin-cell benchmarks to pouch-cell prototypes involves confronting substantial engineering challenges. Therefore, accurately assessing AZIB commercial viability mandates performance validation under realistic electrolyte and loading conditions consistent with pouch-cell configurations, emphasizing the importance of lean electrolyte usage, optimized electrode architectures, and minimal excess zinc. These factors collectively define a significant research gap, underscoring the necessity for intensified efforts in pouch-cell evaluation to better approximate the true commercial potential and limitations of AZIB technologies.

4. SEPARATORS AND CURRENT COLLECTORS

Separator and current collector design plays a supportive but critical role in achieving the long cycle life and interfacial stability required for AZIB commercialization. Traditional glass fiber separators, while widely used, suffer from limitations such as large pore sizes and poor Zn^{2+} transport control that lead to dendrite formation and side reactions. To overcome these issues, recent studies have introduced engineered separators, such as Janus structures and boron nitride-reinforced membranes, which help regulate ion flux and stabilize the electrode−electrolyte interface.[130−132] Similarly, advances in current collector materials, including corrosion-resistant foils and carbon-based three-dimensional (3D) frameworks, have improved Zn deposition uniformity and reduced localized current densities. These innovations contribute to enhanced interfacial performance and cycle stability. However, their practical deployment depends on the development of scalable manufacturing processes and cost-effective integration.[12,133,134]

Despite notable advancements in separator and current collector designs for AZIBs, several commercialization-limiting factors remain unresolved.[130] Functional separators, such as Janus membranes, boron nitride reinforced composites, and charge-selective polymer networks, have shown promise in improving ion selectivity and interfacial stability, yet often involve complex fabrication routes, limited scalability, or high material costs.[131,133,134] Similarly, while carbon-coated or MXene-modified current collectors can suppress dendrite formation and improve electrochemical performance, these materials typically require vacuum deposition or high-temperature processing, limiting their compatibility with large-scale roll-to-roll production. Moreover, conventional copper and stainless-steel foils face corrosion challenges in mildly acidic aqueous electrolytes, raising concerns over long-term durability. To bridge the gap between lab-scale prototypes and commercial deployment, future research must prioritize separator and current collector solutions that balance chemical stability, manufacturability, and cost-effectiveness under realistic cell configurations and cycling conditions.

5. ARTIFICIAL INTELLIGENCE-DRIVEN STRATEGIES FOR AZIBS

Artificial intelligence (AI) techniques have recently been extensively studied for battery commercialization. In particular, AI-based modeling has been successfully implemented for materials discovery, performance prediction and process optimization for battery commercialization. The AI applications for batteries include optimizing electrode and electrolyte formulations, accelerating the identification of degradation pathways, and guiding scale-up strategies. AI-based models could significantly reduce the time and cost needed to search for promising electrode and electrolyte materials. Machine learning (ML) and Neural networks (NN) are used to optimize the synthesis parameters (e.g., Ratio of constituents) of Ni-rich NMC cathode materials, solid and liquid electrolytes to accelerate target specifications more efficiently.[135,136] In addition, AI has also been extensively utilized to facilitate the identification of degradation mechanisms and limitations. Artificial neural networks (ANNs) were employed to search for and predict important electronic properties (e.g., highest occupied molecular orbital−lowest unoccupied molecular orbital (HOMO−LUMO) levels and electron affinities) to understand electrochemical stability that affects battery design

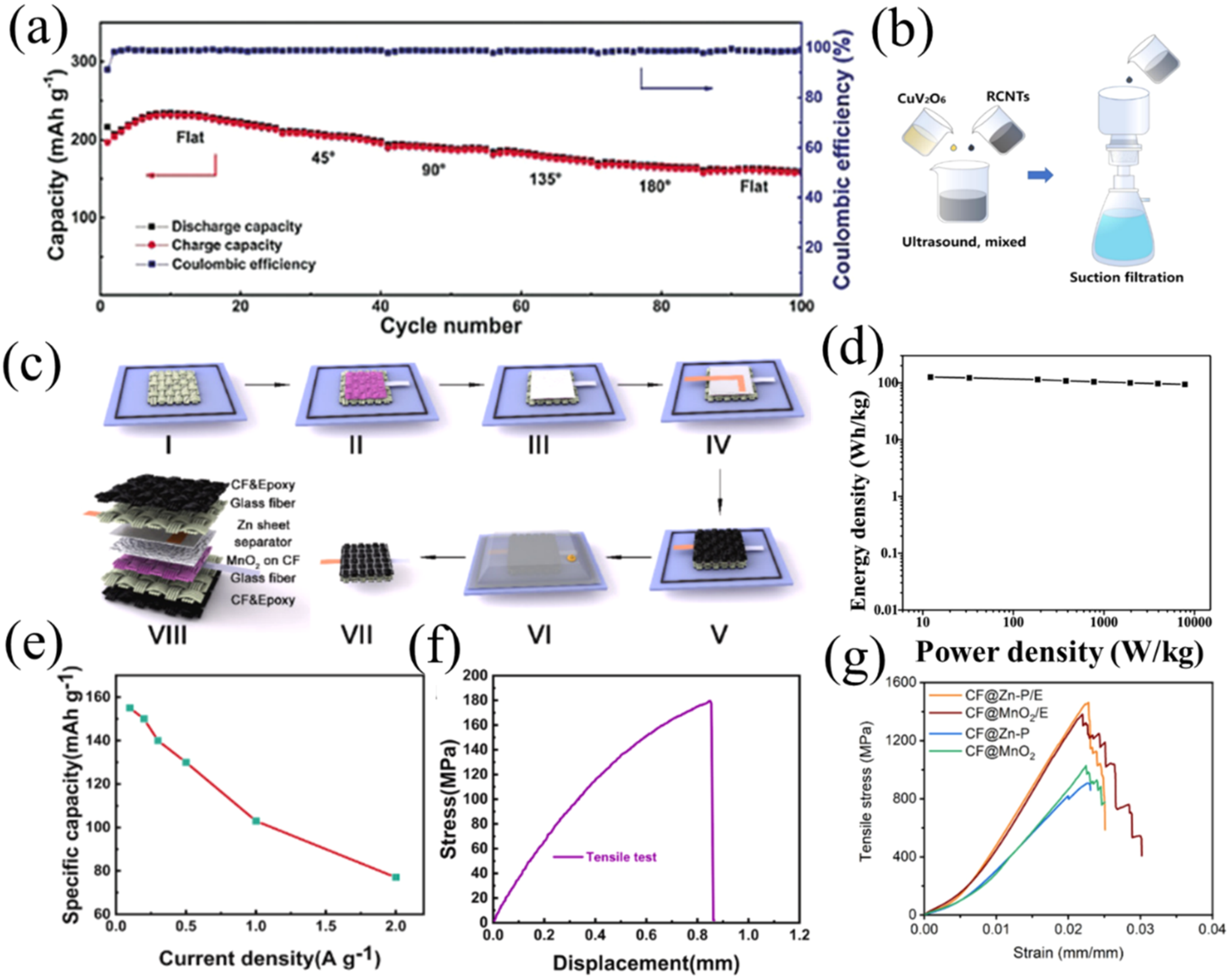

Figure 9. (a) Stability performance of the flexible pyrene-4,5,9,10-tetraone (PTO)-based battery under various bending conditions. Reproduced with permission from ref 141. Copyright 2018, Wiley. (b) Fabrication of the flexible CuV_2O_6 cathode via vacuum filtration. Reproduced with permission from ref 152. Copyright 2021, Elsevier. (c) Schematic of the structural Zn-ion battery with a MnO_2 cathode. Reproduced with permission from ref 158. Copyright 2021, Elsevier. (d) Ragone plot of the Zn-MnO_2 composite. (e) Specific capacity versus current density of carbon-fiber-reinforced ZnSBs and Tensile test results for (f) ZnSB with resin-impregnated carbon fiber. Reproduced with permission from ref 158. Copyright 2021, Elsevier. and (g) high-performance ZnSB with epoxy-impregnated carbon fiber fabric. Reproduced with permission from ref 159. Copyright 2023, The Authors. Preprint available via SSRN.

and durability.[137] Furthermore, AI has contributed to the design of scalable manufacturing processes by combining experimental data with optimization algorithms. Statistical tools from experiments and ML models were integrated to determine the optimal composition of electrode slurries for high-power applications and designed experiments. Identifying material quality and parameters in large-scale production is also essential for battery commercialization and these identifications have been implemented in battery production facilities.[138] It means that data-driven monitoring not only increases throughput but also ensures reliability in cell manufacturing. Therefore, AI can guide scale-up processes and manufacturing optimization in large-scale cell production via feedback loops and digital twins.[139]

6. STRUCTURAL AND FLEXIBLE ZINC-ION BATTERIES: IMPLICATIONS FOR COMMERCIALIZATION

The mechanical properties, particularly flexibility, of AZIBs are essential to their commercialization for structural and wearable devices. Therefore, the traditional rigid structures must be replaced with softer materials to enable flexibility while maintaining good electrochemical performance. Various strategies have been explored to improve the flexibility of cathodes, such as coating active materials on flexible current collectors and directly growing active materials on desired flexible substrates.[140]

Coating active materials on flexible current collectors using a binder and conductive additive is a widely adopted approach.[141−145] For example, Guo et al.[141] fabricated a wearable AZIB based on a pyrene-4,5,9,10-tetraone (PTO) cathode. To fabricate the flexible electrode, they mixed the active material with a conductive additive (Ketjen black) and a binder (poly(tetrafluorethylene), PTFE) and subsequently

Table 6. Comparative Performance Metrics and Structural Trade-Offs in Flexible and Structural Zinc-Ion Batteries

type	energy density	cycle life	mechanical strength	flexibility	implication	refs
structural	205 Wh kg^{-1} (active); 97 Wh kg^{-1} at 2 A g^{-1}	85.3% after 300 cycles at 1 A g^{-1}	tensile: 179.5 MPa; flexural: 229.6 MPa; modulus: ∼4.9 GPa	rigid laminate	lab-validated prototype	158
structural	265 mAh g^{-1} (∼160−200 Wh kg^{-1})	stable under 20 MPa tension	flexural: 268 MPa; modulus: ∼ 4.9 GPa	rigid composite	experimental proof-of-concept	160
structural	181.5 Wh kg^{-1}	88.3% after 100 cycles	strength: 293 MPa	rigid	early stage lab validation	157
structural	7.8 Wh kg^{-1}	90% after 1000 cycles	strength: 206.1 MPa	rigid cathode	material-level demonstration	159
flexible	327.7 Wh kg^{-1} at 135 Wh kg^{-1}	80.7% after 1400 cycles	fiber-shaped; MPa-scale	flexible fiber	advanced laboratory prototype	161
flexible	126 Wh kg^{-1}	88.7% after 1000 cycles	electrodeposition on carbon cloth	bendable fabric	lab-validated with integration potential	162
flexible	54 mAh cm^{-2} areal capacity	stable under bending/twisting	printed on elastomer	flexible, foldable	prototype nearing application trials	163

rolled the mixture on a titanium mesh. Their flexible battery maintained stable capacity even after multiple folding cycles (Figure 9a). However, conductive additives and binders are inactive components that hinder ion and electron transport, which may result in increased electrode polarization and a decrease in energy density depending on the total mass of the entire electrode.[146]

Researchers have also explored in situ growth of active materials on flexible substrates for fabricating flexible cathode electrodes with high electrochemical conductivity and improved mechanical properties (Table 6).[147,148] The most commonly used substrates can be categorized into two main groups: metal-based and carbon-based substrates. Metal-based substrates, including stainless steel (SS) mesh, nitinol wire, titanium mesh, and nickel foil, have been widely employed for their mechanical flexibility. Meanwhile, carbon-based substrates, such as graphene foam, carbon nanotubes (CNTs), and graphite paper, typically exhibit superior shape memory effects compared to their metal-based counterparts. Additionally, carbon-based substrates are generally more cost-effective, abundant, and lightweight, making them a more favorable choice for flexible ZIBs.

Although the primary in situ growth methods can be categorized into hydrothermal[149] synthesis and electrodeposition,[150] vacuum filtration[151,152] has also been used to fabricate flexible electrodes without using binders or conductive additives. For instance, Song et al.[152] developed a freestanding CuV_2O_6/reductive acidified carbon nanotube (CVO/RCNTs) film using vacuum filtration with outstanding flexibility (Figure 9b). The resulting flexible battery exhibited stable electrochemical performance under different bending conditions. However, current flexible cathodes still fall short of the mechanical flexibility required for wearable devices, necessitating further advancements in material design and structural engineering.

Moreover, a new concept in battery technology based on structural battery composites (SBCs) has gained increasing attention due to the high energy efficiency and low environmental impact, with the fabrication procedure schematically shown in Figure 9c. SBCs are multifunctional materials that possess both mechanical strength and electrochemical storage capabilities, enabling operation in diverse applications ranging from electric vehicles to wearable devices.[153] The concept of structural batteries was first introduced by Snyder et al.,[154] who integrated electrochemical storage into composite materials, paving the way for multifunctional energy storage devices. Since then, advancements in laminated, fiber-reinforced, and sandwich-structured batteries have demonstrated the potential for lightweight and durable energy solutions. The global structural battery market is projected to grow up to \$808.24 million by 2032.[155] To make structural batteries practical, one of the most crucial parameters for SBCs is energy density, as it directly reflects their electrochemical performance. For instance, in aviation, one of the potential applications for zinc-based structural batteries (ZnSBs), an energy density in the range of 400−700 Wh kg^{-1} is required to meet practical performance demands.[156] To approach this goal, Chen et al.[157] fabricated a Zn-MnO_2 composite battery via hydrothermal synthesis, reporting a specific capacity of 145.9 mAh g^{-1} and energy density of 126 Wh kg^{-1} with enhanced mechanical durability due to strong adhesion between MnO_2 and carbon fibers (Figure 9d). Similarly, Liu et al.[158] developed carbon-fiber-reinforced ZnSBs with a MnO_2 cathode and Zn anode, achieving an energy density of 205 Wh kg^{-1}, a notable advancement over earlier designs and comparable to those of lithium-ion structural batteries (Figure 9e).

Another important parameter for SBCs is mechanical strength, which allows their use in various applications that demand high mechanical robustness. Although no specific target for mechanical properties exists, fabricating structural batteries with sufficient robustness is essential to applications such as airplanes or vehicles. Significant efforts have been made to improve properties such as tensile strength and Young's modulus. Liu et al.[158] fabricated a ZnSB using impregnated carbon fibers and glass fibers with epoxy resin by using the vacuum infusion method. Their ZnSB showed (Figure 9f) a high tensile strength and Young's modulus of 179.5 MPa and 3.05 GPa, respectively. In a recent study, Liu et al.[159] fabricated a carbon fiber electrode reinforced with epoxy resin, achieving a high tensile strength of 1466 MPa (Figure 9g).

However, enhancing mechanical strength often introduces a trade-off with electrochemical performance, which is an inherent challenge in structural battery design. To achieve high tensile or flexural strength, these systems typically incorporate a significant proportion of mechanically robust yet electrochemically inactive materials, such as carbon fibers, glass fabrics, and epoxy resins. While such components enhance stiffness and load-bearing capacity, they compromise the overall energy density at the composite level by reducing the proportion of active materials. Conversely, increasing the content of active materials content or employing porous and electrochemically favorable architectures may compromise structural integrity and reduce mechanical robustness under applied stress. Therefore, balancing mechanical and electrochemical functions necessitates thoughtful structural design and materials integration. For instance, Liu et al.[158] achieved an electrode-level energy density of 205 Wh kg^{-1} while maintaining a tensile strength of 179.5 MPa by directly integrating MnO_2 into carbon fibers using a vacuum infusion method. This approach highlights the potential of multifunctional materials and engineered interfaces in mitigating the performance trade-off, although further innovations are required to simultaneously enhance energy density and structural robustness at the device level.

Beyond electrochemical and mechanical performance, the manufacturing scalability and structural integration of ZnSBs present substantial challenges to practical deployment. Most currently used fabrication methods, such as hydrothermal growth, electrodeposition, and vacuum-assisted resin infusion, are labor-intensive, batch-oriented, and lack compatibility with high-throughput industrial manufacturing. Scaling these processes requires integration with established composite fabrication techniques, such as automated fiber placement or resin transfer molding,[164] without compromising the structural or electrochemical integrity of the battery. Furthermore, achieving uniform distribution of active materials and ensuring strong interfacial adhesion within multifunctional laminates remain critical for preventing delamination and electrical inconsistency during operation. Integration into load-bearing systems, such as automotive panels or aerospace structures,[158] also necessitates compliance with stringent safety and mechanical standards, which adds complexity to design, testing, and certification. Additionally, the cost and availability of key materials,[34] such as carbon fibers, conductive binders, and engineered cathode components, can influence manufacturing feasibility and scalability. Overcoming these challenges will require a multidisciplinary effort involving materials science, composite engineering, and manufacturing process development to enable reliable, mass-producible ZnSB systems suitable for real-world structural applications.

Despite these advancements, ZnSBs face several challenges that need to be addressed for commercialization. One major issue is zinc dendrite formation,[165] which can lead to short circuits and capacity loss over prolonged cycling. Additionally, MnO_2 cathodes, while promising, exhibit limited conductivity and phase instability, negatively impacting the overall electrochemical performance. Electrolyte design also remains a critical challenge, as traditional liquid and gel-based electrolytes often lack sufficient mechanical reinforcement, leading to potential failures under repeated mechanical stress. To address these limitations, Wang et al.[165] introduced a solid-state Zn^{2+} electrolyte composed of branched aramid nanofibers and poly(ethylene oxide), which effectively suppressed dendrite growth and improved ion transport. Recent innovations focus on enhancing both electrochemical and mechanical performance to bridge the gap between ZnSBs and commercial lithium-ion structural batteries. Strategies such as electrodepositing zinc onto carbon fibers have improved current distribution and reduced local current density, preventing dendrite growth. Additionally, MXene-coated cathodes have shown potential for increasing conductivity and structural stability,[166] making them promising candidates for commercial applications. However, the high cost may be a concern for their commercial deployment.

7. SUMMARY AND OUTLOOK

AZIBs present a promising alternative to conventional LIBs due to their inherent safety, environmental compatibility, and cost-effectiveness. Despite their potential for large-scale energy storage applications, significant challenges hinder their commercialization. Recent research advancements have notably improved AZIB performance through innovative cathode designs, including defect and oxygen vacancy engineering in manganese-based and vanadium-based cathodes. Such modifications effectively mitigate structural degradation, transition metal dissolution, and sluggish zinc-ion diffusion kinetics, which are critical factors limiting energy density and cycle stability. Moreover, electrolyte engineering, particularly the use of advanced additives and novel electrolyte formulations such as water-in-salt and hybrid organic-aqueous systems, has significantly expanded the electrochemical stability window, enabling higher-voltage operations and improved cycling durability. Anode stability remains another critical aspect, with zinc dendrite growth, hydrogen evolution reactions, and corrosive side reactions severely affecting battery performance and longevity. Surface modifications, including coatings and interphase engineering strategies, have demonstrated substantial effectiveness in enhancing zinc plating uniformity and reducing parasitic reactions. Despite these improvements, practical issues such as low zinc utilization ratios and mechanical instabilities under realistic conditions remain key barriers.

Moreover, flexible AZIB designs and SBCs have gained attention for applications in wearable devices and electric vehicles. Efforts to improve mechanical flexibility and multifunctionality are crucial to advancing AZIB technology for these emerging applications. Nonetheless, achieving commercial viability requires addressing cost, scalability, and the compatibility of advanced materials with current manufacturing processes.

Future research efforts are directed toward the systematic development of scalable and cost-effective material synthesis and manufacturing processes to successfully transition AZIB technologies from laboratory innovations to market-ready products. Strategic focus areas include advanced cathode materials with robust structural integrity, optimized electrolyte formulations that balance performance with regulatory compliance, and comprehensive strategies to stabilize zinc metal anodes under practical operational conditions. Successfully addressing these challenges through integrated research efforts will accelerate the realization of commercially viable AZIBs, contributing significantly to sustainable energy storage solutions.

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Notes

The authors declare no competing financial interest.

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■ LIST OF ABBREVIATIONS

AZIB: aqueous zinc-ion battery
CEI: cathode-electrolyte interphase
SEI: solid-electrolyte interphase
ZUR: zinc utilization ratio
HER: hydrogen evolution reaction
ESW: electrochemical stability window
DOD: depth of discharge
DOE: Department of Energy (U.S.)
LDES: long-duration energy storage
LCOS: levelized cost of storage
GCD: galvanostatic charge−discharge
TRL: technology readiness level
MRL: manufacturing readiness level
EC: ethylene carbonate
DMC: dimethyl carbonate
LFP: lithium iron phosphate
NMC: lithium nickel manganese cobalt oxide
NCA: lithium nickel cobalt aluminum oxide
LCO: lithium cobalt oxide
MNC: MnO_2/residual carbon composite
RC: residual carbon
LP: laponite
BT: bentonite
NSF: National Science Foundation
MXene: transition metal carbide/nitride
PBA: prussian blue analogue
ICP: inductively coupled plasma
CV: cyclic voltammetry
XRD: X-ray diffraction
SEM: scanning electron microscopy
XPS: X-ray photoelectron spectroscopy
EIS: electrochemical impedance spectroscopy
DFT: density functional theory
AIMD: ab initio molecular dynamics
FJH: flash Joule heating
PVDF: poly(vinylidene fluoride)
PVA: poly(vinyl alcohol)
DTPA: diethylenetriamine pentaacetate
SPE: solid polymer electrolyte
PEO: poly(ethylene oxide)

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