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Multifunctional Separator Engineering: Unlocking the Potential of Aqueous Zinc-Ion Batteries

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ABSTRACT

Aqueous zinc-ion batteries (AZIBs) have emerged as an idealcandidate for large-scale energy storage due to their inherent safety and low cost. However, key challenges, including Zn dendrite formation, Zn anode corrosion/passivation, hydrogen evolution, and cathodic dissolution, significantly inhibit the development of high-performance AZIBs with long-term durability. Separators, as one of the important components in the AZIBs, show a critical impact on the electrochemical performance. Nevertheless, the systematic understanding of the underlying mechanism of separators improving the battery performance is still unclear. Here, this article focuses on the research progress of AZIBs separators, providing a systematic review of the core role of separators in AZIBs in mechanical integrity and their influence on the other critical components such as anodes, cathodes, and electrolytes. The engineering strategies for functional separators are then analyzed and categorized into matrix modification, functional grafting, organic coating, and inorganic stacking. Notably, this review summarizes recent advances of functional separators based on the substrates, followed by a discussion of the critical challenges and future directions of the research in functional separators for AZIBs, providing theoretical guidelines to advance the Zn battery technologies.

1 | Introduction

The rapid global depletion of nonrenewable energy sources has intensified the demand for alternative clean and sustainable energy systems. Renewable energy sources such as solar, wind, and hydropower have attracted extensive attention. However, their inherent geographical intermittency and uneven distribution necessitate the deployment of efficient and durable energy storage technologies to ensure a stable energy supply [1, 2]. The conventional lithium-ion batteries have been considered as one of the most promising candidates. However, they face significant safety concerns due to flammable organic electrolytes (risking thermal runaway), and their development is often limited by high price and scarcity of lithium [3, 4]. In contrast, aqueous zinc-ion batteries (AZIBs) have emerged as a promising alternative for grid-scale storage. This stems from their intrinsic safety, environmental compatibility, low cost, and the relatively hightheoretical specific capacity (820 mAh g^{-1}) [5–9].

A typical AZIB comprises a cathode, separator, zinc anode, and aqueous electrolyte [10, 11]. During operation, Zn^{2+} ions migrate through the electrolyte-saturated separator, shuttling between the cathode and the anode. The cathode stores ions through intercalation, conversion, or deposition reactions, while the anode undergoes reversible zinc plating and stripping

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[12–14]. Despite their appealing attributes, AZIBs face several intrinsic challenges, including uncontrolled dendritic growth on the Zn anode, parasitic hydrogen evolution [15–19], dissolution and crossover of transition-metal ions from the cathode [20–22], and concentration polarization due to uneven ionic flux [21, 23]. These issues lead to rapid capacity fading [24–26], poor Coulombic efficiency, and reduced battery lifespan [27, 28].

To address these issues, most studies have focused on the optimization of electrodes and electrolytes. For the Zn anode, strategies include developing alloyed Zn anodes and constructing artificial interphase layers to guide uniform Zn deposition and suppress dendrite formation [29–33]. For cathodes, new high-capacity materials and coatings improve structural stability [34–38]. In electrolyte engineering, systems such as concentrated aqueous electrolytes, hybrid ionic liquids, and gel electrolytes are developed to optimize the Zn^{2+} solvation structure, inhibit hydrogen evolution, and improve interfacial compatibility [39, 40]. In contrast, the structure and composition modification of the separators often receive less attention, despite their critical role in AZIBs to influence the transport behavior of Zn^{2+} and the interfacial stability. This oversight is notable because functionalizing the separator offers unique advantages over optimizing other components. Unlike electrode or electrolyte modifications, which often address localized issues, a functional separator provides a holistic and interfacial solution. It can simultaneously regulate ion transport pathways and interfacial electrochemistry between the electrodes, serve as an integrated platform to suppress dendrites, enhance ion transport, and mitigate side reactions, all through a single component. This paradigm shifts the separator's role from passive isolation to active management, presenting a potentially more efficient and cost-effective strategy for performance enhancement. The separators not only physically isolate the electrodes, but also directly determine Zn^{2+} flux distribution, electrolyte wettability, ionic conductivity, and dendrite suppression capability, exerting a profound impact on overall electrochemical performance [41, 42]. Currently, commercial glass fiber separators are most widely used in AZIBs due to their high porosity and excellent hydrophilicity, which enable effective electrolyte uptake [43]. Nonetheless, their large and irregular pore diameters result in nonuniform ionic flux, aggravating localized Zn deposition and dendrite growth [44, 45]. Moreover, their low puncture resistance makes them vulnerable to dendrite penetration [46–48], while their lack of ion-selectivecapability fails to block the shuttling of dissolved transition-metal ions, leading to parasitic reactions at the anodes and severe capacity degradation [49]. These limitations highlight the urgent need to design functional separators that integrate dendrite suppression, ion selectivity, electrolyte regulation, and interfacial stabilization [40, 50].

Therefore, in this review, we provide a comprehensive overview ofseparator engineering strategies for AZIBs. Unlike previous reviews, which have primarily focused on the basic properties and preparation methods of functional separators, this work provides a distinct perspective by elucidating how these components are integral to the interplay between key battery components. We first discuss the significant role of functional separators in addressing key challenges and highlight the principles guiding their design. We then categorize separator optimization strategies into several representative approaches, including matrix modification, functional grafting, organic coating, and inorganic coating/stacking, with emphasis on how each improves ion transport, interfacial chemistry, and mechanical integrity. Furthermore, our review offers the first systematic classification and analysis of recent advances based on substrate types, providing a clear framework for understanding material selection. Finally, we summarize the current challenges and outline future research directions, aiming to bridge the gap between fundamental research and practical applications, and to provide theoretical and practical guidance for the development of high-safety, long-lifetime, and high-energy-density AZIBs.

2 | Functional Roles of Separators in AZIBs

Generally, separators are free-standing porous membranes that support the integrity of the cell and function as a carrier for liquid electrolytes. The use of separators for AZIBs separates the anodes and cathodes from physical contact to avoid electronic connection, while providing sufficient transport pathways for Zn^{2+} shuttling to ionically connect the electrodes. Although often considered passive, separators have the capability to directly influence the electrochemical environment, structural stability, and overall durability of AZIBs. Beyond their fundamental role as ionic conductors and physical barriers, separators can be rationally designed and functionalized to address the unique challenges associated with AZIBs, including Zn dendrite growth, cathode dissolution, side reactions, and mechanical failure [45, 46, 49]. Their multifunctional contributions play a vital role in the critical parts of AZIBs, particularly in terms of mechanical integrity, electrolyte properties, anode behaviors, and cathode activities (Figure 1).

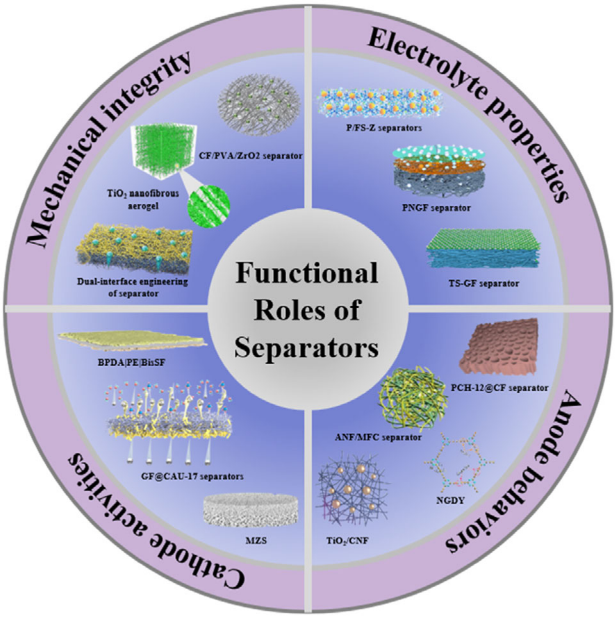

FIGURE 1 | Schematic diagram of functional roles of separators in AZIBs.

2.1 | Mechanical Integrity

As aqueous liquid electrolytes are generally used in AZIBs, which cannot directly function as a physical barrier between anodes and cathodes, the most common role of separators for AZIBs is to support the mechanical integrity of the cells [46–48]. Therefore, separators are required to have excellent dimension stability, which should be able to withstand internal stresses generated during battery operation. That includes repeated Zn stripping and plating, electrolyte swelling, parasitic reactions, and even dendrite formation. Therefore, the separators used in AZIBs are often flexible and resilient instead of rigid and fragile in order to accommodate volume fluctuations of >50% during the long-term cycling. Poor mechanical strength often leads to internal short circuits and thermal runaway, eventually resulting in battery failure. To overcome the challenges, recent studies have emphasized the reinforcement of the conventional separators (e.g., glassfiber, cellulose, polypropylene) through engineering strategies, including incorporating inorganic fillers [51–53], introducing high crosslinking [49, 54–57], and surface coating [58, 59]. For instance, Yao et al. designed a novel inorganic/organic hybrid separator, denoted as P/FS-Z, which consists of a polytetrafluoroethylene (PTFE) matrix, fumed silica (FS) filler, and zinc salt [46]. It is characterized by high mechanical strength, high modulus (Figure 2a), and a balanced hydrophilic-lipophilic design that resists dendrite penetration. Zhang et al. developed a cellulose/PVA/ZrO_2 (CF/PVA/ZrO_2) composite separator with an asymmetric structure [47]. One side features a functional layer rich in ZrO_2, designed to precisely control ion transport and deposition behavior, while the other side incorporates highly cross-linked cellulose, providing mechanical support and ion conduction capability (Figure 2b). By combining mechanical blocking and electric field regulation, it effectively suppresses the growth of zinc dendrites. Kim et al. coated the surface of a glass fiber (GF) separator with a polyvinylidene fluoride (PVDF) layer to suppress structural deformation caused by prolonged exposure to the electrolyte [48]. These strategies not only improve the mechanical integrity of the cells, but also contribute to the enhancement of the electrochemical performance, which are indispensable for advancing the safety, reliability, and scalability of next-generation AZIBs.

2.2 | Electrolyte Properties

In addition to supporting the mechanical integrity of the batteries, separators can also contribute to the electrolyte properties in AZIBs, through regulating electrolyte uptake and retention, ionic transport, and interfacial stability. In general, an ideal separator for AZIBs should possess high liquid absorption capability and good wettability to ensure continuous ion transport channels, enabling sufficient Zn^{2+} diffusion with minimal internal resistance. The conventional separators suffer from poor affinity with aqueous electrolytes, leading to uneven ion flux and polarization [62, 63]. Thus, they are often engineered through morphology modification to increase porosity and change pore size distribution [64–66], surface functionalization to enhance electrolyte affinity and reduce interfacial resistance [48, 67], and composition incorporation to enhance transport pathways and stability [46, 53, 68, 69], which enhances the properties of electrolytes and facilitates the performance of AZIBs. For instance, Zhu et al. significantly enhanced the electrolyte affinity by grafting acrylic acid (AA) onto a low-cost and highly ion-conductive yet hydrophobic polypropylene (PP) separator, resulting in a separator with hydrophobic/hydrophilic domains (denoted as PP-g-AA) [60]. In this structure, the polypropylene (PP) polymer backbone partially blocks water molecules and prevents side reactions, while the carboxyl functional groups from acrylic acid (AA) enables the adsorption of zinc ions, ensures uniform the electric field distribution, and establishes a dynamic transport chain for zinc ions, facilitating uniform zinc deposition while mitigating dendrite formation and side reactions (Figure 2c). Furthermore, the chemical composition of the separators may also affect the local environment in the electrolyte, which may provide additional functionalities such as ionic gradients and cation/ion selectivity to facilitate the battery reactions [70]. Ma et al. proposed an ultra-thin and lightweight separator featuring an asymmetric functional coating [61]. On the face-to-cathode side, the BPDA coating inhibited the shuttle effect and the spontaneous dissolution of vanadium species. This reinforced the cathode structure and mitigated the self-discharge rate by 40%. On the face-to-anode side, the BisSF coating guided a uniform and rapid Zn^{2+} flux, inducing preferential Zn deposition along the (002) crystal plane while repelling $SO_4{}^{2-}$ and promoting desolvation (Figure 2d). Hence, dendrites, HER and micro-corrosion side reactions were effectively inhibited, even in high-throughput scenarios (20, 20 mAh cm^{-2}). By synergistically modulating electrolyte uptake, ion selectivity, and transport kinetics, separators play a critical role in dictating the electrolyte environment and electrochemical performance of AZIBs.

2.3 | Anode Behaviors

Zn metal anodes have been widely used in AZIBs due to their abundance and low cost. However, there are several challenges associated with the use of Zn metal anodes, including dendrite formation, parasitic reactions, and low interfacial stability [16–19, 15]. These may not only compromise Coulombic efficiency, but also decrease the cell safety, leading to internal short circuits and termination of the batteries [24–26]. On the other hand, the design of separators significantly affects the zinc deposition pattern and the stability of the zinc negative electrode. The pore structures of the separators often play a significant role in the distribution of Zn^{2+} flux. In particular, the uniformly distributed small pores can promote lateral ion diffusion and prevent dendrite nucleation. Therefore, regulating the pore structure of the separators is crucial to inhibit the formation of Zn dendrites at the Zn metal electrodes [65, 71]. In addition, reasonable adjustment of the electric field on the surface of the separator may also help to promote the uniform distribution of Zn^{2+}, while creating negatively charged surfaces through functional grafting (e.g., sulfonic acid group [72–74]) can further regulate the ion flux through electrostatic repulsion. For instance, Hong et al. developed a cation-exchange membrane of perfluorosulfonic acid (PFSA) coated on the Zn surface to regulate the Zn plating/stripping behavior [75]. Importantly, the PFSA film with abundant sulfonic acid groups could simultaneously block the access of $SO_4{}^{2-}$ and H_2O, accelerate the Zn^{2+} ion transport kinetics, and uniformize the electrical and Zn^{2+} ion concentration field on the Zn surface (Figure 3a), thus achieving a highly reversible Zn plating/stripping process with corrosion-free and dendrite

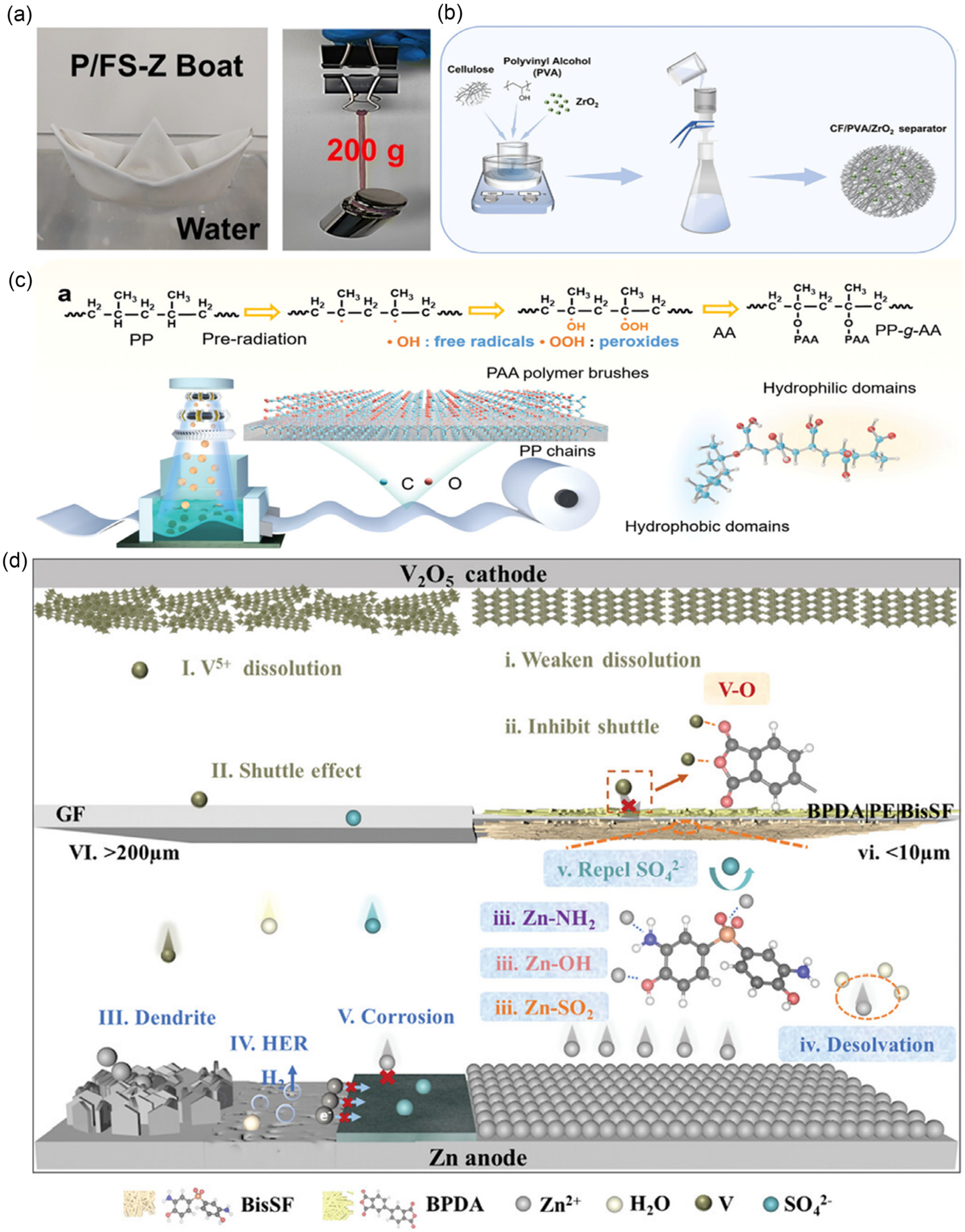

FIGURE 2 | (a) Demonstration of flexibility and hydrophilic–hydrophobic balance characteristics. Reproduced with permission [46]. Copyright 2023, Royal Society of Chemistry. (b) Schematic illustration of the fabrication of $CF/PVA/ZrO_2$ separators. Reproduced with permission [47]. Copyright 2024, Wiley-VCH GmbH. (c) Schematic illustration of the preparation of the PP-g-AA separator. Reproduced with permission [60]. Copyright 2024, Wiley-VCH GmbH. (d) A schematic illustration depicting the synergistic effects of the Janus-type separator strategy. Reproduced with permission [61]. Copyright 2024, Wiley-VCH GmbH.

free behavior. Another major challenge to inhibit the behavior of Zn metal anodes is their limited stability. Corrosion inevitably occurs at the interface of Zn anodes, leading to parasitic reactions including passivation of Zn metal and hydrogen evolution reactions [16–19, 15]. Therefore, optimization of separators by introducing different functional groups [60, 78–80] and surface functional coating [67, 68] can adjust the electric field distribution and buffer pH fluctuations, which can effectively inhibit the hydrogen evolution and the accumulation of byproducts on the surface of negative electrodes. In addition, adjusting the electrolyte absorption capacity of the separator through composition engineering is also an effective strategy to improve the anode stability. For instance, an ANF/MFC [76] composite separator with hydrophilic/hydrophobic structures was developed to

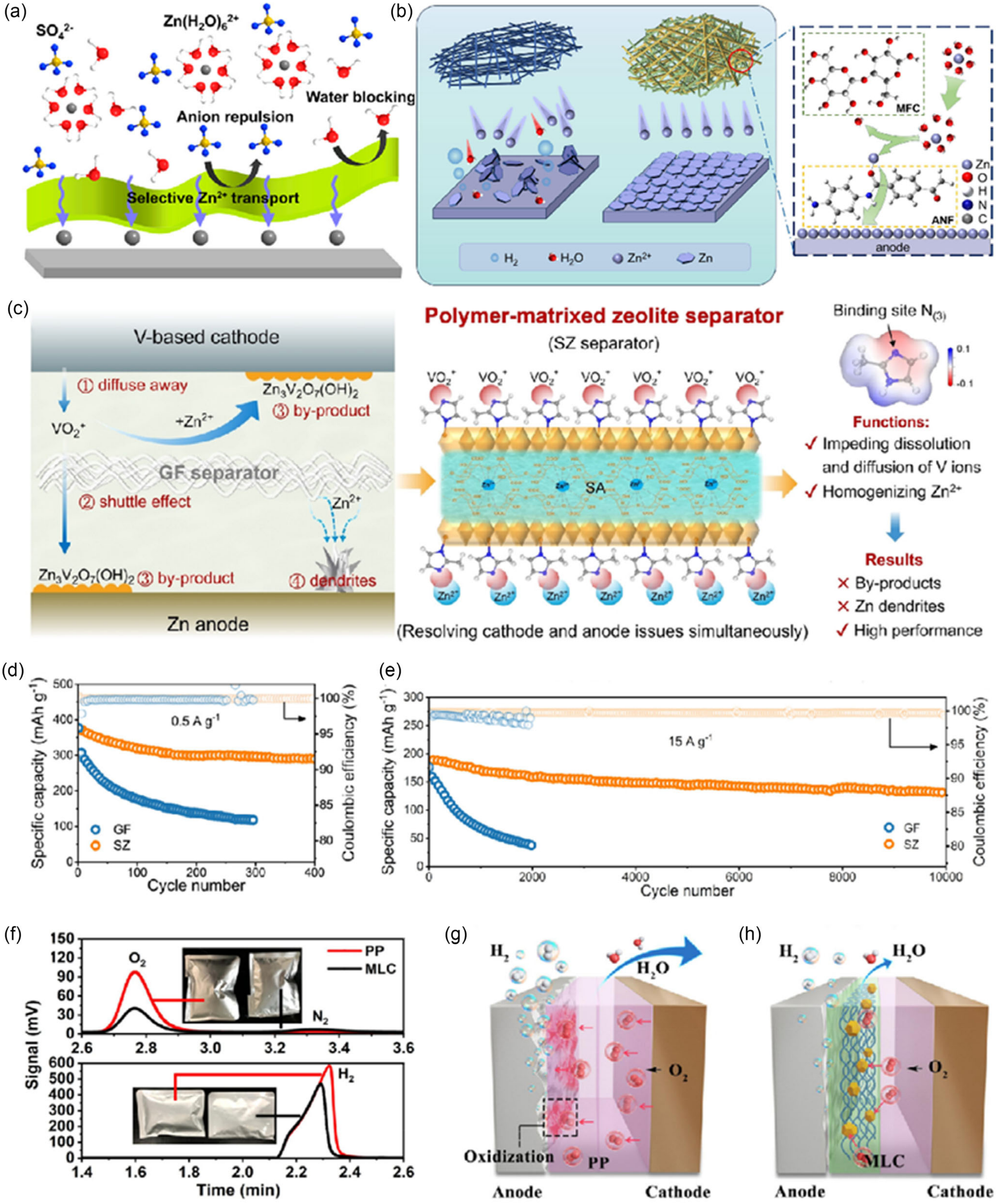

FIGURE 3 | (a) Schematic illustration of the Zn deposition process on the Zn metal anode with the protection of the PFSA film. Reproduced with permission [15]. Copyright 2022, American Chemical Society. (b) The schematic diagram for the performance-improvement mechanism of the ANF/MFC separator compared with the GF separator. Reproduced with permission [76]. Copyright 2025, Elsevier Ltd. (c) Schematic diagram of the challenges Zn/V cells face and the working mechanism of as-prepared separator. Cycling performance of the full cells at a current density of (d) 0.5 A g^{-1} and (e) 5 A g^{-1}. Reproduced with permission [49]. Copyright 2023, Wiley-VCH GmbH. (f) Gas chromatograms for PP and MLC cells after storage test and floating charge. Schematic diagram of various side reactions during storage and floating charge of (g) PP and (h) MLC cells, respectively. Reproduced with permission [77]. Copyright 2022, Elsevier B.V.

utilize the synergistic effect of hydrophobic aramid nanofibers (ANF) and hydrophilic microfibrillated cellulose (MFC) which created localized dehydrated zones at the interface of Zn anode with an improved ratio of Zn^{2+} /H_2O (Figure 3b) [76]. As a result, the hydrogen evolution and surface passivation were effectively inhibited, leading to the significantly improved battery performance, with cycling stability of over 3800 h at 1 mA cm^{-2} in the Zn//Zn symmetric configurations.

2.4 | Cathode Activities

Separators also play a critical role in stabilizing the cathodes of AZIBs, which often suffer from dissolution and degradation of active materials from low structural stability. This may also lead to the shuttle effect that ions of active substances (e.g., V^{4+}/V^{5+}, Mn^{2+} generated from Mn^{3+} disproportionation) dissolved on the cathodes to reach the negative side, which may lead to negative electrode toxicity and electrochemical degradation [49, 81–83]. The separator, as a physical barrier, can either prevent the dissolution or the migration of the ion couples through the modification with the inorganic coating to achieve a balance of the ion accessibility with the inhibition of anodic degradation. A typical example is a polymer matrix zeolite separator (SZ) which was synthesized by in situ growth of zeolite material (ZIF) on sodium alginate (SA) polymer membrane [49]. The separator acted as an effective ion-blocking layer to prevent V leaching and shuttling from the cathode, while significantly inhibiting the formation of by-products and Zn dendrites (Figure 3c). The SZ cell was stable for over 10 000 cycles at 15 A g^{-1} (Figure 3d,e). Furthermore, developing functional separators also contributes to maintaining local electrolyte composition adjacent to the cathodes, buffering pH fluctuations to suppress parasitic reactions such as oxygen evolution reactions which are detrimental to cathode stability. For instance, Zhao et al. proposed a novel “multifunctional layered conductive” (MLC) separator [77]. The separator has the enhancements in conductivity and zinc adsorption, which lead to the even current density and zinc deposition, respectively. Besides, the separator has improved water retention capacity, conductive agent with high hydrogen evolution, and block effect of oxygen (Figure 3f–h). Consequently, various side reactions and their corresponding negative effects have been suppressed, including evaporation of electrolyte, HER, and OER. It is worth noting that the modification of separators for improving electrolyte properties (e.g., wettability, ionic transport) can also have positive impact on the cathode activity, by minimizing concentration gradients and enhancing cathode reaction kinetics, especially at high current densities [48]. These results verify the pivotal role of separators for AZIBs with improved electrochemical performance.

3 | Engineering Strategies of Separators for AZIBs

The separators have been demonstrated to play a role far more than a passive barrier in the AZIBs. As the center of the cell structure, separators often show direct impact on the electrochemical behavior of the integrated system, regulating the ion transport, interfacial reactions, and mechanical stability. Advanced separators significantly improve the performance of aqueous zinc-ion batteries by simultaneously regulating ion transport, desolvation, and interface stability [84]. In terms of ion transport, functional separators guide uniform zinc ion distribution through electrostatic effects or nanochannels, suppressing zinc dendrite formation [68, 85]. During desolvation, polar groups on the separator surface weaken the solvation shell of hydrated zinc ions, reducing the energy barrier for deposition and minimizing side reactions [48, 82, 86]. For interface stability, dense functional layers serve as physical barriers against dendrite penetration while stabilizing the interface by accommodating volume changes and homogenizing the electric field [86, 87]. These three mechanisms are interconnected: uniform ion transport enables even deposition, efficient desolvation reduces side reactions, and a stable interface ensures long-term cycling stability, all working together to enhance overall battery performance (Table 1). However, the conventional separators, including cellulose membranes, glass fiber scaffolds, and polyolefin films, often face severe limitations to fulfill these missions in AZIBs. Therefore, to address these challenges, separators are required to be functionalized through various engineering strategies to ensure safe operations and enable long cycle lives. Here we categorize the engineering strategies of separators for AZIBs into four main domains: matrix modification, functional grafting, organic coating, and inorganic stacking (Figure 4).

3.1 | Matrix Modification

Among the engineering strategies, matrix modification of separators is often considered as the direct way to change the intrinsic properties to facilitate the battery performance. Instead of introducing the secondary supportive species (e.g., functional groups and surface coating), the matrix modification focuses on tailoring the framework and structural properties of the separators, which have a significant impact on the anodes, cathodes, and the overall system. The morphology of the separator framework, as a typical example, can be tuned to possess different porous structures with high surface areas. Generally, small pores may act as physical barriers against dendrite penetration on Zn anodes and material dissolution on cathodes [45, 102, 103], while large pores may facilitate uniform electrolyte uptake and rapid Zn^{2+} transport. For instance, Jiao et al. fabricated a cellulose fiber PAN/CA/HNTs composite separator exhibiting hydrophilicity, thermal stability, and enhanced ion transport properties [65]. The separator, produced using NIPS technology, features a gradient pore structure (Figure 5a). The gradient pore architecture of the separator offers storage capacity for electrolytes, thereby enhancing Zn^{2+} transport and kinetics. The composition of the separators also plays an important role in regulating the properties. Applying polymer blending with hydrophilic polymers in the framework often enhances the electrolyte affinity and ionic conductivity [60], while compositing with inorganic fillers within the main structure benefits the mechanical strength and thermal resistance [51–53, 106]. Furthermore, increasing the crosslinking degrees within the polymeric separators can also improve dimension stability, contributing to the suppressed swelling and structural integrity during long-term cycling [91]. For instance, Cheng et al. used polyacrylonitrile (PAN) and zwitterionic surfactant (sulfobetaine methacrylate, SBMA) as precursors to fabricate a modified PAN nanofiber separator via electrospinning [74].

TABLE 1 | Advantages and disadvantages of separator modification strategies for AZIBs.

Strategy	Mechanism	Key advantages	Main challenges	References
Matrix modification	Intrinsically designing the separator's physical structure.	- Excellent structural integrity and uniformity. - Avoids risks of layer delamination. - Maintains inherent mechanical strength of the base material	- Limited functional versatility (primarily physical). - Heavily dependent on the base material's properties. - Low flexibility for post-design chemical adjustments.	[65, 74, 88–91]
Functional grafting	Functional grafting precisely introduces functional groups at the molecular level.	- Stable and permanent functionality via covalent bonds. - Molecular-level precision for targeted functions (e.g., ion steering). - Does not significantly alter bulk structure or thickness.	- Complex and costly processing (e.g., plasma, UV). - Difficult to scale up and ensure uniform grafting.	[48, 60, 64, 92–94]
Organic coating	Applying a functional layer onto the separator surface.	- Wide variety of available polymers for diverse functions. - Simple, scalable, and industry-friendly coating processes. - Highly compatible with existing battery manufacturing.	- Potential poor adhesion, leading to coating delamination. - May increase ionic resistance and hinder rate capability. - Requires precise control over coating thickness and uniformity.	[83, 95–97]
Inorganic stacking	Constructing a layer of inorganic nanomaterials on the separator.	- Superior mechanical strength to block dendrite penetration. - Enhanced thermal stability and safety. - Nanochannels can regulate and sieve ion transport.	- Inherent brittleness can lead to crack formation. - Poor compatibility with electrolyte may increase interface resistance. - Challenges in forming uniform, defect-free layers.	[58, 59, 98–101]

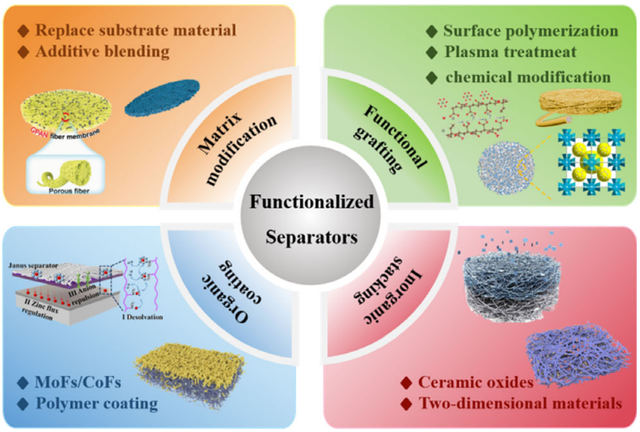

FIGURE 4 | Schematic diagram of engineering strategies of separators for AZIBs.

The incorporation of the zwitterionic surfactant SBMA effectively modifies the viscosity and enables a more homogeneous distribution of the electrostatic field, thereby optimizing the electric field characteristics during the electrospinning process and significantly enhancing the mechanical properties of the fibers, as shown in Figure 5b. Consequently, a Zn//Zn symmetric cell assembled with PAN@SBMA separator shows a prolonged cycle life of 1700 h, and the Zn//PAN@SBMA//$NH_4V_4O_{10}$ full cells deliver higher specific capacity (236 mAh g^{-1}) and excellent long-term durability of 84.2% capacity retention after 2000 cycles at 5A g^{-1}, which is significantly better than cells assembled with GF-D separator. It can be concluded that the modification of the separator matrix can enhance the activities of the battery components and maintain a high mechanical strength against short-circuiting and capacity fading, thereby addressing the unique challenges of AZIBs.

In summary, matrix modification optimizes the separator's intrinsic architecture to achieve high structural integrity and mechanical strength, while offering limited functionality and being highly dependent on the base material. It is highly scalable using established industrial processes and cost-efficient, leveraging common polymer formulations for significant performance gains at minimal extra cost.

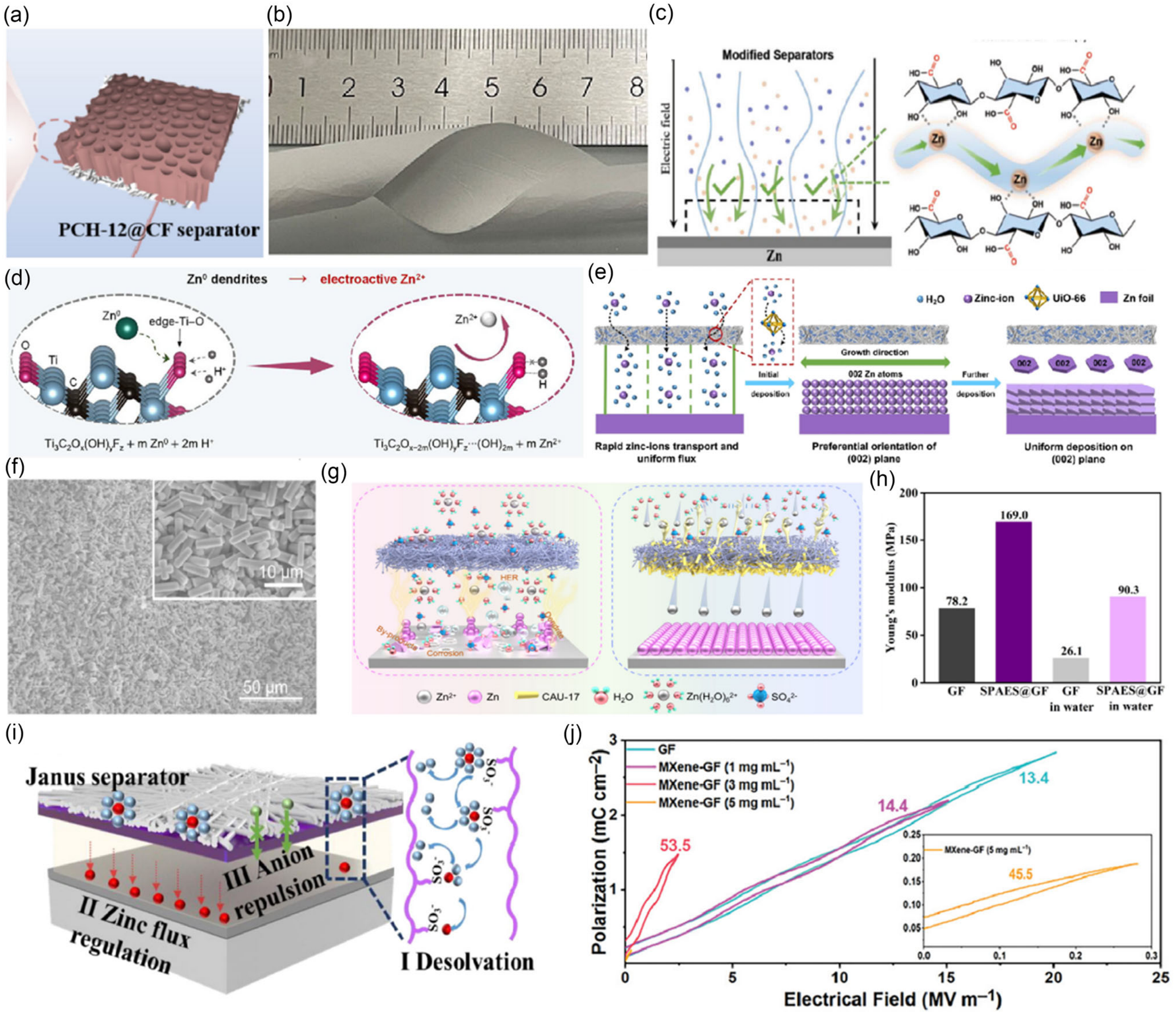

FIGURE 5 | (a) Morphology of PCH-12@CF separator. Reproduced with permission [65]. Copyright 2025, Elsevier B.V. (b) The flexible ability of the PAN-based separator. Reproduced with permission [74]. Copyright 2024, Wiley-VCH GmbH. (c) Mechanism of uniform zinc deposition with modified separators (CCM). Reproduced with permission [107]. Copyright 2023, Wiley-VCH GmbH. (d) The spontaneous redox process between Zn^0 and the Ti-O$_e$ functional group on MesoTi_3C_2. Reproduced with permission [104]. Copyright 2023, American Chemical Society. (e) Mechanism comparison of the deposition processes for zinc anodes using UiO-66-GF-2.2. Reproduced with permission [105]. Copyright 2023, the Author(s). (f) SEM and magnified (inset) images of the GF@CAU-17 separator. (g) Schematics of Zn deposition with GF (left) and GF@CAU-17 separators (right). Reproduced with permission [83]. Copyright 2023, Wiley-VCH GmbH. (h) Mechanical strength of different separators. (i) The schematics of Zn deposition with the designed Janus separator. Reproduced with permission [97]. Copyright 2023, Elsevier. (j) Polarization-electric field (P-E) loops of GF and MXene-GF. Reproduced with permission [101]. Copyright 2022, Wiley-VCH GmbH.

3.2 | Functional Grafting

Functional grafting is an effective strategy to endow separators with tailored surface chemistry and ion selectivity in AZIBs. This approach usually involves applying specific functional moieties on the separator matrix to provide functionalities such as regulating Zn^{2+} transport, suppressing parasitic reactions, and stabilizing electrode interfaces. In general, introducing hydrophilic moieties such as —OH, —NH_2, and —COOH can improve the electrolyte affinity and enhance homogeneous Zn^{2+} flux, and grafting ionic groups such as anions (e.g., —SO_3^-, —COO^-) and cations (e.g., imidazolium, pyridinium) can regulate the surface ionic gradients to facilitate Zn plating and stripping while suppressing the cathode dissolution [48, 60, 68, 79]. The engineering of separators through functional grafting can be achieved through surface polymerization [60], plasma treatment [48], or chemical modification [92, 105], enabling precise control over functional group density and distribution. For instance, Yu et al. synthesized ultra-long bioproduced polymer nanofiber-derived separators CCM (the carbonylation modification cellulose membranes) with high Zn^{2+} transfer number by oxidizing the polymer units of biosynthesized cellulose CM and introducing carbonylated side groups. The results showed that the introduction of carboxylate groups on the separator enhanced the Zn^{2+} transfer capacity due to the loosened interaction between the polymer backbone and Zn, leading to the timely replenishment of

Zn^{2+} to the ion depletion zone near the anode, which resulted in uniform Zn deposition (Figure 5c). In addition to the common functional groups, redox-active moieties can be introduced to serve as secondary electron mediators, suppressing the cathode dissolution and stabilizing electrochemical performance. Bu et al. proposed a new proactive dendrite-digesting chemistry via the $MesoTi_3C_2$-wrapped PP separator [104]. The $MesoTi_3C_2$, with active edge sites, can effectively revive the dead Zn^0 dendrites into electroactive Zn^{2+} ions (Figure 5d), thereby enhancing the longevity of aqueous ZABs. The formation of $MesoTi_3C_2$ with evenly distributed in-plane mesopores relies on the high redox potential difference-driven homogeneous oxidation of both edge and basal planes of MXenes. Furthermore, in situ growth of crystalline porous materials with abundant functional ligands, such as MOFs (metal–organic frameworks) [78, 79, 108] and COFs (covalent organic frameworks) [72, 92], on the separator can also provide additional functionalities to guide the diffusion of Zn^{2+} and induce the uniform deposition of Zn, owing to their capability to adjust the solvation structure and reduce the activity of water. Song et al. reported the in situ growth of Zr-based metal-organic backbone material (UiO-66) on the commercial glass fiber separators to obtain UiO-66-GF [105]. UiO-66-GF effectively enhanced the charge carrier transport capacity and exhibited preferential orientation of the (002) crystalline surface (Figure 5e), which was conducive to the improvement of corrosion resistance and the realization of dendrite-free Zn deposition. (Metal–Organic Frameworks Functionalized Separators for Robust Aqueous Zinc-Ion Batteries) The rational design and application of functional-group-grafted separators provide significant improvement in the electrochemical performance, making them a key strategy for constructing durable AZIBs.

In conclusion, functional grafting precisely introduces functional groups at the molecular level through chemical bonding with permanent modification advantages, though it involves complex processes and faces challenges in scaling up. Consequently, its scalability is moderate, and the cost-effectiveness is also moderate, making it more suitable for high-value applications.

3.3 | Organic Coating

Organic coating is another widely adopted strategy to introduce multifunctionality to separators in AZIBs. However, the preparation of organic coating appears relatively easy compared to that of functional grafting, since the coating strategy does not require covalent bonds to the separator frameworks, making this method more feasible and widely used. Generally, applying organic coating modification achieves precise synergy between ion transport modulation and interfacial stabilization on the surface of the separator. Similar to functional grafting, organic coating usually relies on the types of functional moieties on the organic backbones to maintain strong interactions to Zn^{2+} or repel anions, which facilitates the ion migration to achieve cation-selectivity. For instance, Shao et al. fabricated a GF@CAU-17 separator by depositing CAU-17 (a Bi-based MOF) onto a glass fiber (GF) membrane via vacuum filtration (Figure 5f) [83]. As shown in Figure 5g, CAU-17 is enriched with carboxyl groups that effectively repel harmful SO_4^{2-} ions, blocking their entry, and simultaneously disrupting the pristine HBs of free H_2O molecules to reconstruct the HBs, thereby fundamentally impairing parasitic side reactions and undesirable by-products. Moreover, the superior zincophilic properties of CAU-17 facilitate interactions with Zn^{2+} ions, promoting the desolvation of $[Zn(H_2O)_6]^{2+}$, which creates a self-expedited Zn^{2+} ion pump that dynamically maintains an even and fast Zn^{2+} flux, eliminating concentration gradients and enabling homogeneous Zn^{2+} deposition. Based on the intrinsic properties of the organic materials used in the coating, the resultant separators often exhibit corresponding properties in the AZIBs. Coating Zn-affiliated polymer, such as polyacrylic acid (PAA) and polyethyleneimine (PEI), accelerates ion migration through a high density of charged groups [109–112]. Using biomimetic adhesive materials, including polydopamine (PDA) [67] and gelatin, reveals an enhanced interfacial compatibility by utilizing catechol/amino groups. And developing multifunctional copolymers coating can significantly enhance mechanical strength and ionic conductivity. A typical example is the functional Janus separator constructed by Sun et al. through coating a dense layer of sulfonated poly(arylene ether sulfone) (SPAES) on a commercial glass fiber separator, which significantly enhances the mechanical strength of the separator (Figure 5h) [97]. As shown in Figure 5i, the sulfonic acid groups on the surface of the dense SPAES layer form a double electric layer that repels anions and reduces parasitic reactions, while the glass fiber substrate with a porous structure maintains sufficient electrolyte absorbance, enabling an ultra-long cycle life of more than 2000 h operation at a face capacity of 1 mAh cm^{-1}, which is a 23-fold enhancement over the use of the pristine glass fiber separator. Another commonly used organic coating material is conductive polymers, including polypyrrole (PPy), polyaniline (PANI) [113], and poly(3,4-ethylenedioxythiophene) (PEDOT) [114]. These polymers usually display high electronic conductivity, ideal for improving electron transfer and creating uniform Zn^{2+} transport channels, thereby suppressing dendrite growth and enhancing plating/stripping reversibility. In addition, these materials may sometimes exhibit redox activity [104], extending their role from passive barriers to active participants in the anodic and cathodic reactions. Through their versatility in chemistry, conductivity, and flexibility, organic coatings transform conventional separators into multifunctional interfaces capable of stabilizing both anode and cathode processes, ultimately improving the cycle life and efficiency of AZIBs.

To summarize, the organic coating strategy integrates multiple functions by applying a organic functional layer onto the separator surface to balance functional diversity and process feasibility, yet encounters issues with interfacial adhesion and ion transport resistance. In addition, due to the low cost of raw materials like polymers and biomass derivatives, its scalability is good, and cost-effectiveness is high.

3.4 | Inorganic Stacking

Inorganic coating has emerged as a powerful strategy to enhance the stability, mechanical robustness, and ion transport properties of separators in AZIBs. Different from organic coating, by integrating a dense layer of inorganic materials onto polymer or fibrous membranes, separators gain synergistic enhancement of physical confinement and chemical modulation, improving resistance against dendrite penetration and undesirable side reactions. Ceramic oxides, such as Al_2O_3, TiO_2, and SiO_2, are often used based on their high mechanical strength, thermal stability, and polar surfaces that enhance electrolyte wettability [115–118]. These coatings homogenize Zn^{2+} ion flux, reduce local current density, and effectively block Zn dendrites from short-circuiting the cell. Two-dimensional materials, including MXenes [119, 120], BN [121, 122], MoS_2 [123], and graphenes [124], are known for their unique 2D architectures, which improve the structural integrity while providing additional functionalities such as conductivity, electrostatic interactions, and parasitic reaction suppression [58, 125, 126]. Among them, MXene shows potential for coating applications due to its unique microstructure and excellent electrical conductivity. Su et al. prepared a Janus separator (MXene-GF) by spraying $Ti_3C_2T_X$ MXene on the commercial glass fiber [101]. The surface of this separator was enriched with polar groups, which provided good electrolyte wettability and high ionic conductivity, homogenizing the local current distribution and promoting Zn nucleation. Figure 5j shows that MXene-GF has a tunable dielectric constant (optimized value of 53.5), which generated a directional electric field to accelerate the transport of Zn^{2+} and repel anions, resulting in a homogeneous distribution of the electric field on the Zn anode to achieve dendrite-free growth. In general, inorganic coatings transform separators into mechanically reinforced and ion-selective barriers, which simultaneously protect the anode and cathode interfaces while prolonging the cycle life of AZIBs.

In summary, the inorganic stacking strategy deposits a nanomaterial layer on the separator, granting exceptional mechanical strength, thermal stability, and electrochemical regulation, but suffers from inherent brittleness and poor interfacial compatibility. These issues, coupled with increased weight and thickness, result in limited scalability, low cost-effectiveness, and safety risks from detachment, hindering its widespread application.

4 | Functional Separators for AZIBs

It has been established that separators, especially the functional ones after engineering, play a pivotal role in the electrochemical performance of AZIBs. Separators not only provide ionic conductive pathways, but also influence electrolyte uptake, Zn^{2+} transport, dendrite suppression, and blocking dissolved transition-metal species. It has been revealed that the intrinsic properties of the substrate material may dictate electrochemical performance, mechanical robustness, and compatibility with aqueous electrolytes. Therefore, we summarize the functional separators of AZIBs into three categories: glass fiber separators, cellulose separators, and polymer separators.

4.1 | Glass Fiber Separators

Glass fiber (GF) has become a widely used separator material in AZIBs due to its low electrical conductivity, low cost, high mechanical strength, suitable porosity, chemical inertness, and good compatibility with aqueous electrolytes. Their loose fibrous structure ensures abundant electrolyte uptake and facile Zn^{2+} transport, making them attractive for fundamental studies and baseline comparisons [45]. However, this separator also has significant defects. The internal pore size is large and unevenly distributed, resulting in a low ion transfer rate and non-uniform paths [45, 103]. It can easily lead to the rapid formation and spread of Zn dendrites, which ultimately penetrate the weak mechanical strength of the separator and lead to short circuits within the battery. Furthermore, due to the lack of ion species on the glass fiber substrate, it is difficult to regulate the ion selectivity and facilitate Zn^{2+} transport [61, 70]. Therefore, modification or reconstruction of glass fiber separators is an effective way to improve the electrochemical performance of AZIBs.

One approach to solving the issue of large pore size and uneven distribution is to reconfigure the separator matrix. Wu et al. reconstructed glass fiber separators by preparing polyacrylonitrile/silica (PAN/SiO_2) composites through electrospinning combined with roll pressing [127]. The resulting membranes exhibited a dense cross-stacked nanofibrillar structure, with SiO_2 nanoparticles uniformly distributed on the surface (Figure 6a). With silica as the main reinforcing component at an optimal loading of 30 wt%, the PSO-30 separator demonstrated a mechanical strength of 7.10 MPa, which was ~37 times higher than pristine glass fiber (0.168 MPa), effectively resisting dendrite penetration (Figure 6b). Moreover, the nanofiber structure enhanced ionic conductivity, accelerating Zn deposition kinetics. The symmetric cells assembled with PSO-30 separators achieved a long cycle life of over 1150 h at 0.5 mA cm^{-2}, with improved energy density and capacity retention in full cells.

In addition to structural reconstruction, chemical modification of glass fiber can further regulate ion transport. Liu et al. introduced a supramolecular modification strategy, fabricating GF@SM separators by grafting supramolecules into the pores of commercial GF using ultrasonic filtration (Figure 6c) [64]. The supramolecules effectively filled the large voids, yielding a more uniform pore structure that facilitated Zn^{2+} migration. More importantly, supramolecular groups such as carbonyl, triazinyl, and amino moieties provided abundant Zn-friendly binding sites, guiding ordered Zn^{2+} deposition and preventing dendrite growth. This functional grafting not only enhanced interfacial stability, but also significantly improved the electrochemical performance of the modified separator in AZIBs.

Surface coating is a simpler yet effective approach, and organic modifications have been used to tune the Zn^{2+} flux distribution. For instance, Wang et al. deposited phthalocyanine (Pc) molecules on the anode-facing side of glass fiber separators via vacuum filtration to construct Pc-GF separators [128]. During Zn deposition, Pc coordinated with Zn to form zinc phthalocyanine (ZnPc), generating an electrostatic field layer that balanced the interfacial electric field and homogenized Zn^{2+} flux (Figure 6d,e). Finite element simulations confirmed the ability of the Pc coating to regulate ion transport, suppress dendrites, and prevent direct contact between the Zn surface and electrolyte. This design also enhanced electrode corrosion resistance and suppressed water-induced parasitic reactions, yielding stable and dendrite-free Zn anodes.

Inorganic coatings provide an additional route to reinforce mechanical and interfacial stability. For example, Liang et al. modified glass fiber separators by depositing BaTiO3 (BTO) nanoparticles onto the anode-facing side via vacuum filtration [59]. The nanoparticles filled surface gaps, producing a

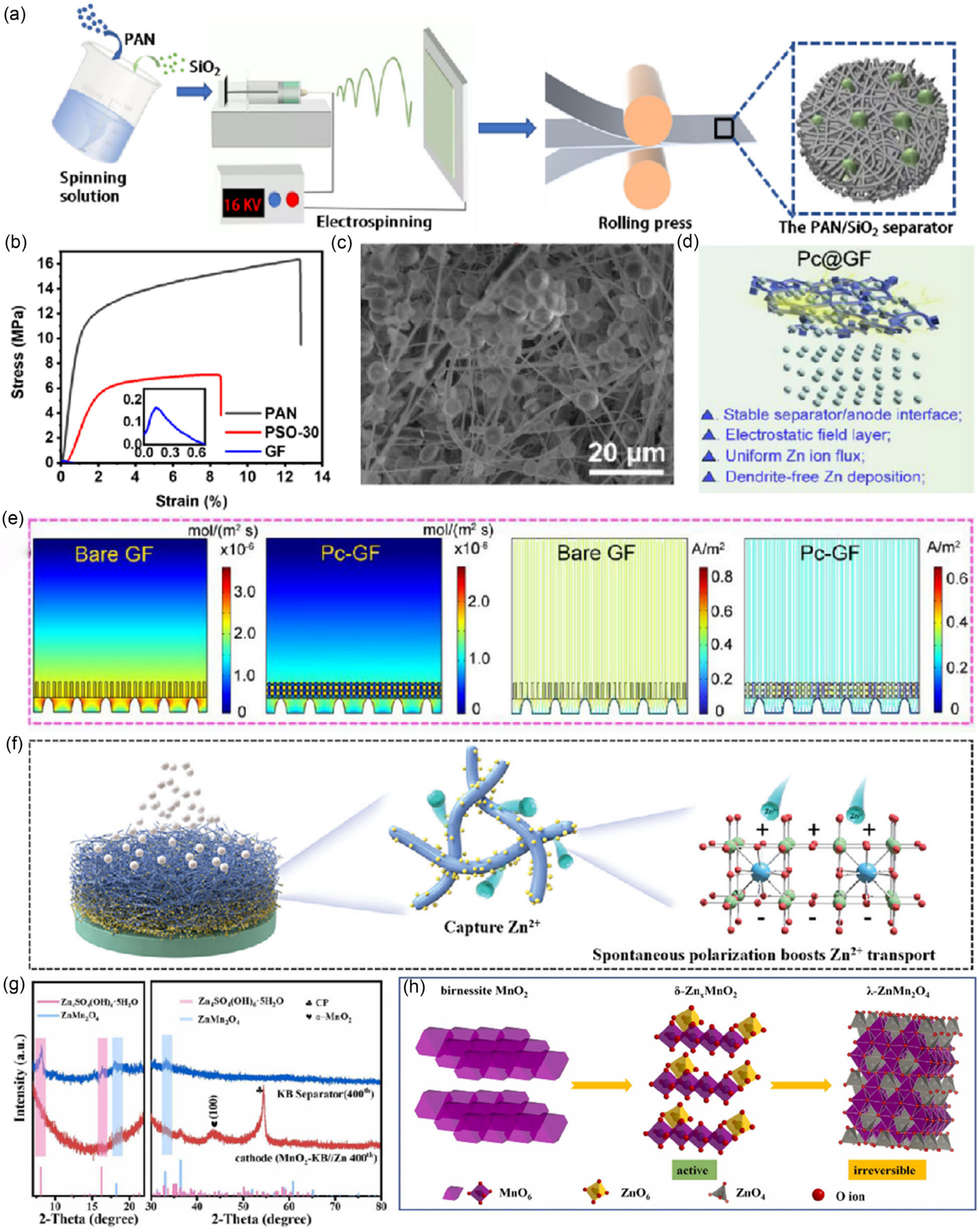

FIGURE 6 | (a) Schematic illustration of preparation of the PAN/SiO$_2$ separators. (b) Tensile curves of the PAN, PSO-30, and GF separators. Reproduced with permission [127]. Copyright 2024, Elsevier B.V. (c) SEM image of composite GF separator with supramolecules. Reproduced with permission [64]. Copyright 2021, Elsevier B.V. (d) Schematic illustration of the Zn deposition assisted by PC-GF separator (e) Simulated results of the Zn ion concentration and electric field distribution using different separators. Reproduced with permission [128]. Copyright 2025, Wiley-VCH GmbH. (f) Schematic separator of enhanced mechanism for Zn^{2+} transport via dual-interface engineering. Reproduced with permission [59]. Copyright 2022, Wiley-VCH GmbH. (g) XRD spectra of the KB separator of the MnO$_2$-KB//Zn battery in the fully charged state after 400 cycles at 2 A g^{-1}. (h) Schematic illustration of the crystal structure transitions that in the battery with the KB separator during cycling. Reproduced with permission [129]. Copyright 2023, Elsevier B.V.

smoother interface and uniform ion transport. Benefiting from the spontaneous polarization effect and high zincophilicity of BTO, Zn^{2+} migration was accelerated, and dendrite-free deposition was achieved (Figure 6f). The modified Zn anode showed highly reversible plating/stripping at 10 mA cm^{-2} with a cumulative capacity of 9500 mAh cm^{-2}, while Zn-MnO_2 full cells maintained 108 mAh g^{-1} capacity after 1800 cycles at 1 A g^{-1}. The glass fiber separators can also be tailored to improve cathode utilization. Huang et al. coated glass fiber separators with a conductive carbon black (KB) layer on the cathode-facing side (Figure 6g) [129]. This modification enabled preferential deposition of Zn hydroxy-sulfates (ZHS) on the separator instead of the anode, lowering impedance and improving ion transport. Furthermore, the KB layer could electrooxidize dissolved Mn^{2+} back into active layered manganese oxides (Figure 6h), mitigating cathode degradation and enhancing full-cell cycling stability.

In summary, while glass fiber separators are widely used, their intrinsic structural flaws necessitate modification. Strategies such as structural reinforcement, supramolecular pore-filling, and functional coatings have been developed to enhance their mechanical strength, regulate ion flux, and suppress side reactions, significantly improving the cycling stability of AZIBs.

4.2 | Cellulose Separators

Cellulose, a natural macromolecule composed of glucose units, is one of the most abundant renewable polymers on earth [45, 130]. It shows desirable characteristics such as high mechanical strength, good flexibility, hydrophilicity, electrical insulation, and biodegradability, making it a promising substrate for developing next-generation separators in AZIBs [45, 90, 130, 131]. Nevertheless, cellulose separators also face certain limitations. Their natural hydrophilicity, while beneficial for electrolyte uptake, can lead to excessive swelling, dimensional instability, and reduced mechanical strength in long-term operation [130, 132]. In addition, their relatively poor thermal stability compared to synthetic polymers limits their application under high-power or elevated-temperature conditions [131, 133]. Without further modification, cellulose membranes may also lack sufficient ion selectivity, failing to fully suppress dissolved transition metal ions crossover or parasitic reactions [54, 107, 118]. These drawbacks highlight the need for continued engineering of cellulose-based separators, especially via hybridization, surface functionalization, and structural reinforcement, to fully unlock their potential in practical AZIBs systems.

Reconstructing cellulose substrates provides a straightforward route to improve pore architecture and mechanical robustness. For instance, Zhou et al. fabricated cotton cellulose (CF) separators through a simple filtration process, thereby creating a new substrate with dense and uniform nanopores, abundant hydroxyl groups, and superior mechanical properties (Figure 7a,b) [45]. The uniform porous structure effectively suppressed side reactions and dendrite formation. As a result, the Zn symmetric cells with CF separators remained stable for a cumulative plating capacity of 1000 mAh cm^{-2} and tolerated deep cycling at high areal capacities up to 20 mAh cm^{-2}. Furthermore, full cells coupling with MnO_2 exhibited markedly enhanced rate performance and cycling stability compared to cells with conventional separators, demonstrating the potential of cellulose as a reliable and cost-effective separator matrix.

Introducing functional groups onto cellulose backbones allows for precise tuning of ion transport and selectivity. A typical example is that Zhou et al. developed a sulfonated cellulose (CF-SO_3) separator featuring low thickness, high mechanical strength, and excellent ionic conductivity (Figure 7c) [134]. The grafted $-SO_3^-$ groups provided dual benefits, including electrostatic repulsion against SO_4^{2-} anions, and strong coordination with Zn^{2+} cations. This design inhibited anion migration, suppressed lateral Zn^{2+} diffusion, and promoted desolvation of hydrated Zn^{2+} ions, thereby enabling uniform vertical deposition (Figure 7d,e). Benefiting from these features, Zn symmetric cells with CF-SO_3 separators achieved dendrite-free plating at 4 mAh cm^{-2} for 1200 h and still maintained activity for 400 h at a large discharge depth of 68.3%. This work highlights functional grafting as an effective approach to boost both interfacial stability and long-term cycling durability.

In addition to modifying cellulose-based separators using functional group branching, surface coatings offer a simpler and scalable strategy to enhance separator performance. Yang et al. modified cellulose separators with two-dimensional porous g-C_3N_4 nanosheets via drop-coating (Figure 7f) [58]. The g-C_3N_4 layer, rich in uniformly distributed nanopores, ensured homogeneous Zn^{2+} flux and guided dendrite-free Zn deposition. Meanwhile, the coating increased the mechanical strength of the separator, preventing short circuits and structural collapse during cycling. The synergistic effect of mechanical reinforcement and ion flux regulation significantly improved electrochemical performance, while the g-C_3N_4 layer maintained structural stability even after extended operation, demonstrating excellent durability.

Beyond single-component coatings, cellulose can be engineered into multifunctional composite designs by incorporating inorganic materials. Zheng et al. prepared an ultrathin Janus separator composed of bacterial cellulose (BC) nanofibers integrated with Ag nanowires (AgNWs) through a simple filtration process (Figure 7g,h) [54]. The BC framework, rich in hydroxyl groups, provided excellent wettability and a uniform nanoscale pore structure, while the AgNWs served as ion pumps and homogeneous nucleation sites. The laser confocal microscopy revealed that Zn anodes paired with pristine BC separators developed rough surfaces with dendritic aggregates, whereas those with Janus separators exhibited smooth, dense, dendrite-free morphologies (Figure 7i,j). Moreover, the AgNWs network enhanced thermal conductivity, enabling uniform heat distribution and eliminating local hotspots (Figure 7k,l), which is crucial for stable Zn electrodeposition. This Janus design endowed the separator with superior mechanical, thermal, and electrochemical properties, pointing to its promise for next-generation high-performance AZIBs.

In summary, cellulose separators offer a sustainable and mechanically robust alternative. Through structural engineering, functional group grafting, and composite design, their limitations can be overcome to achieve uniform pore structures, enhanced

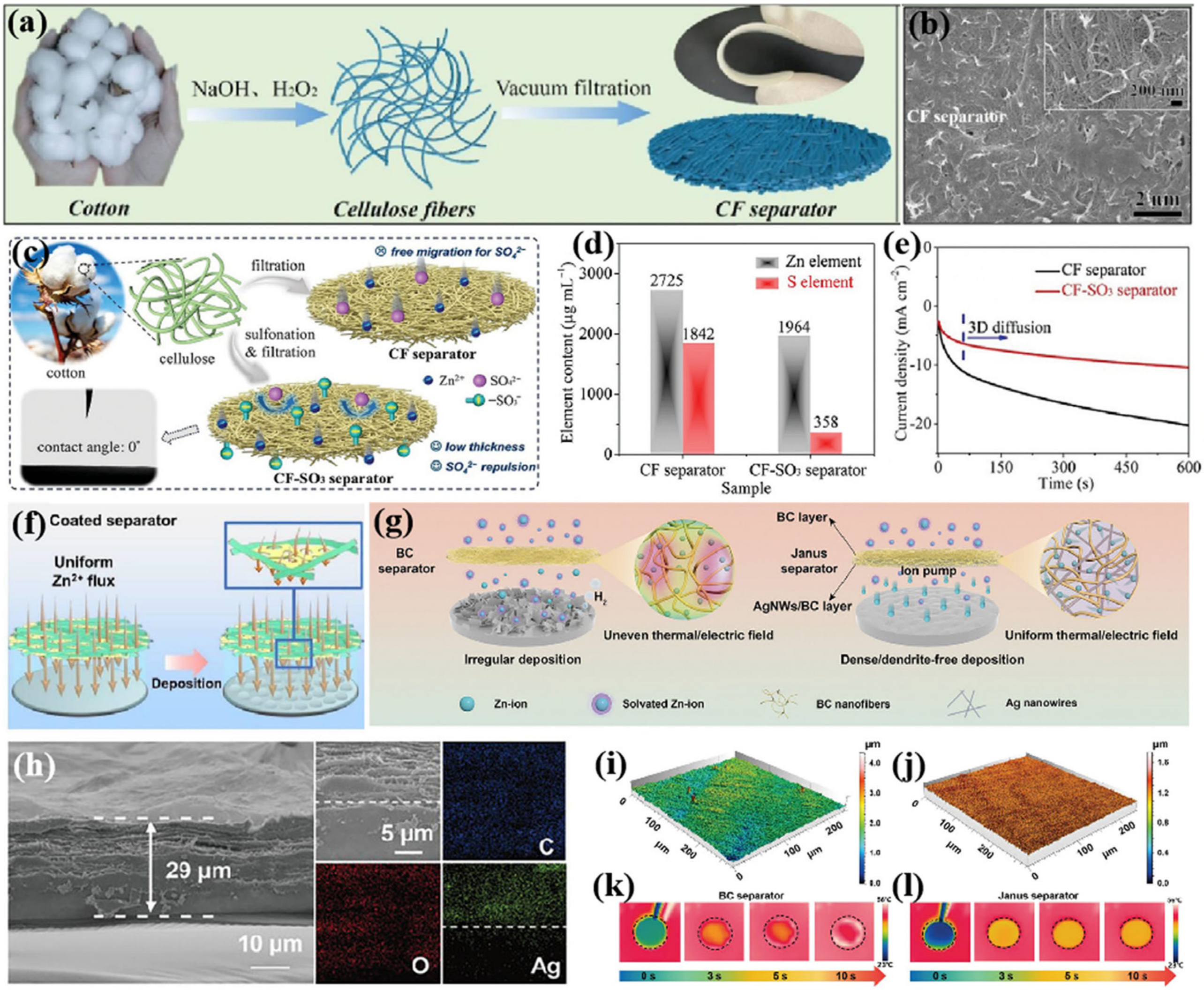

FIGURE 7 | (a) Schematic illustration of the fabrication of CF separator. (b) SEM image of CF separator. Reproduced with permission [45]. Copyright 2022, Elsevier B.V. (c) Schematic separator of the preparation processes. (d) Concentrations of Zn and S elements in the part B of the cell after the permeation tests. (e) CA curves of the Zn//Zn cells with CF or CF-SO_3 separator. Reproduced with permission [134]. Copyright 2024, Wiley-VCH GmbH. (f) Schematic separators of zinc deposition with g-C_3N_4 coated separator. Reproduced with permission [58]. Copyright 2022, Elsevier B.V. (g) Schematic illustration of function mechanism of BC and Janus separators. (h) The cross-sectional SEM images with corresponding elemental mapping of Janus separator. Laser confocal microscopy 3D topographic images of zinc deposition were obtained after 100 cycles at 1 and 1 mA h cm^{-2} with (i) BC and (j) Janus separators. Infrared thermal images of (k) BC and (l) Janus separators upon heating. Reproduced with permission [54]. Copyright 2023, Wiley-VCH GmbH.

ion selectivity, and guided Zn deposition, making them promising for next-generation AZIBs.

4.3 | Polymer Separators

Polymer-based separators provide a versatile platform for optimizing the performance of AZIBs owing to their customizable structures and multifunctional properties. On one hand, polymers offer inherent mechanical toughness, chemical inertness, and thermal stability, which contribute to battery safety and cycling durability [135, 136]. On the other hand, functional groups in polymer backbones (e.g., carboxyl, amine, nitrile) can coordinate with Zn^{2+}, promoting directional transport and uniform deposition [57, 137, 138]. However, the intrinsic hydrophobicity of many polymers limits electrolyte wettability and ionic conductivity, making the engineering strategies of functionalization highly desirable [60].

Polyacrylonitrile (PAN) is among the most widely investigated polymer substrates [139, 140]. Fang et al. fabricated electrospun PAN-based separators with a three-dimensional long-range ordered structure, replacing the inhomogeneous pore distribution of conventional glass fiber separators (Figure 8a) [141]. The nitrile groups in PAN established N-Zn coordination bonds (bond energy –1.8 eV), guiding Zn^{2+} transport along well-defined pathways and reducing interfacial concentration gradients (Figure 8b). This regulation enabled uniform nucleation and dendrite-free Zn deposition, with the deposited layer exhibiting an optimally oriented (101) crystalline surface, which significantly enhanced cycling stability. In addition, PAN suppressed the diffusion of dissolved Mn^{2+}/V^{n+} ions, mitigating cathode

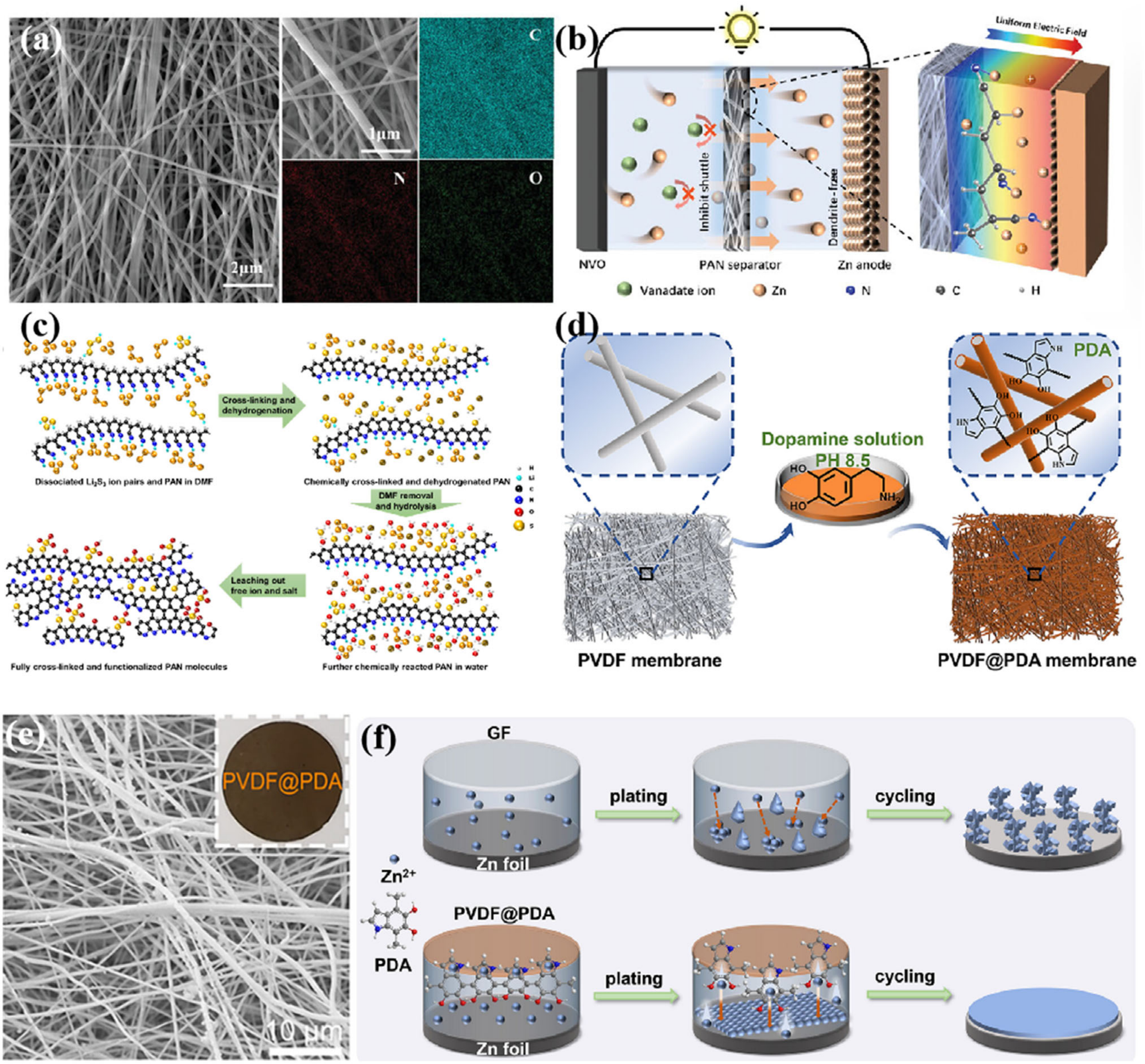

FIGURE 8 | (a) SEM images of PAN separator. (b) The working mechanism of PAN separator in AZIBs. Reproduced with permission [141]. Copyright 2021, Wiley-VCH GmbH. (c) Proposed synthesis route for the novel PAN-S membrane. Reproduced with permission [44]. Copyright 2018, American Chemical Society. (d) Schematic illustration of the fabrication process of the PVDF@PDA separator. (e) SEM images of PVDF@PDA separator. Inset: the photographs of corresponding separators. (f) Schematic illustration of Zn deposition with GF and PVDF@PDA separators. Reproduced with permission [67]. Copyright 2022, the Author(s).

dissolution and by-product formation. As the result, Zn-MnO_2 full cells with PAN separators achieved 84.3% capacity retention after 1000 cycles, which is substantially higher than that of cells with unmodified GF separators (57.8%).

Grafting functional groups onto PAN further enhances electrolyte compatibility and ion selectivity. For instance, Lee et al. prepared PAN-based cation-exchange membranes with improved hydrophilicity and mechanical properties, with the Li_2S_3-assisted crosslinking strategy to introduce sulfonic acid groups (—SO_3H) into the polymer backbone to yield PAN-S membranes (Figure 8c) [44]. The —SO_3H moieties improved wettability, enabled selective Zn^{2+} transport, and promoted desolvation of hydrated ions, leading to dendrite-free electrodeposition. These thin and mechanically robust membranes also blocked dissolved Mn and V species, thereby reducing capacity fading from parasitic reactions. The multifunctionality of grafted PAN membranes highlights the promise of chemical modification in improving separator performance.

Surface functionalization of hydrophobic polymers, particularly PVDF, is an important strategy to enhance electrolyte affinity. Liu et al. coated PVDF membranes with a thin hydrophilic polydopamine (PDA) layer, forming PVDF@PDA separators (Figure 8d,e) [67]. The PDA coating introduced abundant —OH and —NH groups, which coordinated with Zn^{2+} through Zn—O and Zn—N bonds, thereby homogenizing ion flux and suppressing dendritic growth (Figure 8f). As a result, Zn/$NH_4V_4O_{10}$

full cells with PVDF@PDA membranes exhibited excellent rate capability and cycling stability, retaining 92.3% capacity after 1000 cycles at 5 A g^{-1}. This simple organic coating strategy offers a scalable approach to overcome the hydrophobic limitations of PVDF-based membranes.

Hybridizing polymers with inorganic fillers improve both mechanical and electrochemical properties. Likitaporn et al. fabricated PAN/bio-based polyurethane (PU)/$Ti_3C_2T_x$ MXene composite membranes via electrospinning, where Ti_3C_2 MXene nanosheets were embedded within the polymer fibers to form spindle-shaped structures [42]. The incorporation of MXene significantly enhanced electrolyte uptake, ionic conductivity, and charge–discharge stability in symmetric Zn cells. Furthermore, the composite membranes maintained structural and dimensional integrity even after heat treatment, combining flexibility with high performance for durable AZIBs operation.

In summary, polymer separators provide a highly customizable platform. By designing fibrous structures, grafting functional groups, and forming composites, their properties can be tailored to homogenize ion transport, improve interfacial stability, and block cathode dissolution, highlighting their key role in advancing AZIB performance.

5 | Summary and Outlook

In this review, recent advances in separator design for AZIBs have been systematically examined. We first discussed the functional roles of separators, emphasizing their influence on ion transport, dendrite suppression, and interfacial stability. We then summarized major engineering strategies, including matrix modification, functional grafting, organic coating, and inorganic stacking, followed by further categorizing separators based on substrate materials, such as glass fiber, cellulose, and polymers. Recent studies demonstrate that rational separator design can substantially enhance Zn anode reversibility, suppress cathode dissolution, and improve long-term cycling performance.

Despite these promising developments, significant challenges remain before separator technologies can meet the requirements of practical AZIBs applications. Based on our assessment, three critical areas demand focused attention.

5.1 | Commercialization Hurdles

Most reported functional separators have been evaluated in laboratory-scale coin cells, which often fail to capture the complexities of commercial pouch-cell operation. At larger scales, parasitic hydrogen evolution, severe dendrite growth, and interfacial instability become more pronounced, raising doubts about the scalability of current solutions. Moreover, widely used commercial separators such as glass fiber and Nafion remain too costly for the low-price targets of grid-scale storage [103]. Developing low-cost, abundant, and biodegradable alternatives, such as cellulose, chitosan, or other biomass-derived membranes, offers a sustainable path forward, provided their stability and selectivity can be adequately engineered.

5.2 | Underdeveloped Improvement Methods

Among current engineering strategies, functional group grafting has shown superior effectiveness by directly tailoring ion transport and interfacial chemistry. However, scaling these approaches is nontrivial. Backbone reconstruction and composite weaving typically require high technological input, while surface coatings may delaminate or degrade under long-term cycling. Furthermore, multifunctionalization often necessitates multistep synthesis, which complicates cost-efficient production and hinders commercialization. Future work should focus on integrated, scalable fabrication methods, such as one-step electrospinning, in situ polymerization, or green chemical grafting, that can combine multiple functions (dendrite suppression, ion selectivity, mechanical reinforcement) in a single process.

5.3 | Untapped Dual-Interface Benefits

Separators inherently sit between the anode and cathode, yet most designs focus on alleviating challenges at only one interface. A promising direction is the development of Janus separators or asymmetric multilayer membranes, where one side regulates Zn^{2+} deposition while the other side blocks transition-metal crossover or buffers cathode dissolution. The key challenge is to maintain thinness (<50 μm), flexibility, and low cost while ensuring durability during extended cycling. Future studies should also explore dual-interface functionalization coupled with structural optimization (e.g., gradient porosity, thermal conductivity tuning) to exploit the full potential of separators in simultaneously stabilizing both electrodes.

5.4 | Toward a New Era of Smart Separators

Based on a systematic review of existing separator modification strategies, the development of next-generation separators for aqueous zinc-ion batteries will focus on smart separator systems with self-healing capabilities and multi-stimulus responsiveness. These separators can evolve from traditional static barriers into dynamic management systems: their self-healing properties effectively repair microdamage formed during cycling, while their intelligent responses to external stimuli (temperature, pH, potential) provide active protection functions such as overload protection and real-time suppression of side reactions, thereby significantly enhancing the safety margins and service life of batteries.

To achieve this goal, future research needs to deeply explore the intrinsic relationship between the microstructure of separators and their functional properties. The focus should be on elucidating how the geometric characteristics of pore structures (size, distribution, connectivity) regulate ion transport kinetics, interface stability, and intelligent response behaviors. Such an in-depth mechanistic understanding will provide theoretical guidance for the precise design of separator structures, promoting the transition of smart separators from laboratory concepts to practical applications and laying a solid foundation for the development of next-generation high-performance aqueous zinc-ion batteries.

Looking ahead, multifunctional, low-cost, and scalable separator designs are critical to unlocking the full potential of AZIBs. Advances in materials science, such as biomass-derived nanofibers, two-dimensional nanomaterials, and redox-active polymers, combined with innovative architectures like Janus structures, hybrid composites, and self-healing membranes, will shape the next generation of separators. Continued collaboration between fundamental research and applied engineering will be essential to overcome scalability barriers and deliver safe, durable, and commercially viable AZIB systems for large-scale energy storage.

Author Contributions

Chen Qian: conceptualization (lead); writing – original draft (lead). **Limeng Sun:** conceptualization (equal); writing – original draft (equal); writing – review and editing (equal). **Li Gao:** writing – original draft (supporting); writing – review and editing (supporting). **Chao Yuan:** writing – original draft (supporting); writing – review and editing (supporting). **Bin Yu:** writing – original draft (supporting); writing – review and editing (supporting). **Pengpeng Zhang:** writing – original draft (supporting); writing – review and editing (supporting). **Meiliang Chen:** writing – original draft (supporting); writing – review and editing (supporting). **Hong Gao:** writing – original draft (supporting); writing – review and editing (supporting). **Yufei Zhao:** supervision (lead); writing – original draft (supporting); writing – review and editing (supporting). **Hao Liu:** supervision (supporting); writing – original draft (supporting); writing – review and editing (supporting). **Guoxiu Wang:** supervision (supporting); writing – original draft (supporting); writing – review and editing (supporting). **Jinqiang Zhang:** conceptualization (lead); supervision (lead); writing – original draft (supporting); writing – review and editing (lead).

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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