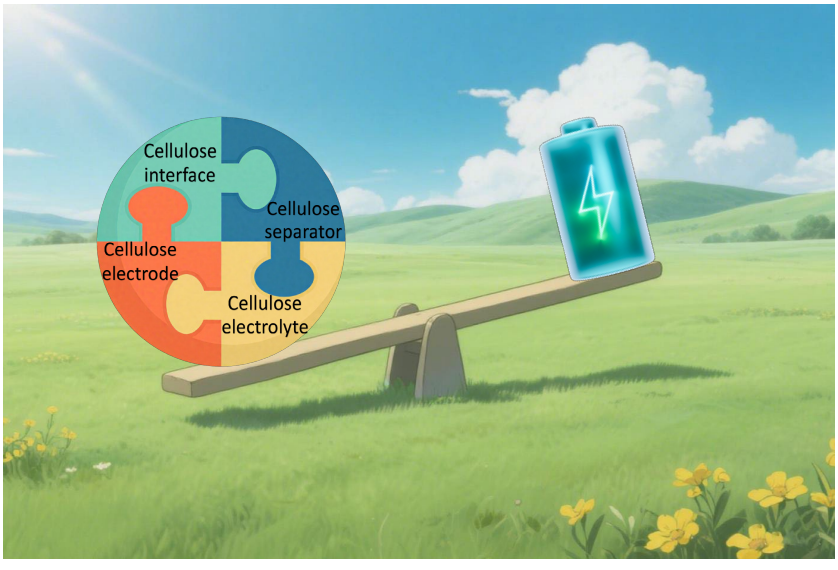
Energy Materials and Devices

Review

The transformative potential of cellulose in energy storage systems

Longjun Chang, Zhaoxin Li, Zhiyuan Yan, and Qing Li ✉

Graphical Abstract



Cellulose-based materials are attracting growing interest in the development of advanced energy storage systems, owing to their intrinsic sustainability, tunable physicochemical properties, and structural versatility. This review provides a critical overview of how these beneficial properties align with the demands of key components—including electrode, the solid electrolyte interphase, separator, and electrolyte—with the aim of accelerating progress toward sustainable cellulose-based energy storage solutions.

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The transformative potential of cellulose in energy storage systems

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ABSTRACT

Cellulose-based materials have attracted growing interest in the development of advanced energy storage systems owing to their intrinsic sustainability, tunable physicochemical properties, and structural versatility. This review systematically summarizes the key features of cellulose from the perspectives of synthesis, physicochemical characteristics, and structural design, highlighting its unique functionality and adaptability. Furthermore, the roles of cellulose in four critical battery components, i.e., electrode, solid electrolyte interphase, separator, and electrolyte, are comprehensively discussed, emphasizing the properties aligning with the specific requirements of each component. Finally, potential research directions are proposed to guide future development. This review provides a comprehensive framework for understanding the transformative potential of cellulose in sustainable electrochemical energy storage systems as well as a guideline for future studies.

KEYWORDS

cellulose, electrode, solid electrolyte interphase, separator, electrolyte

1 Introduction

Rechargeable batteries have profoundly transformed modern life, from powering smartphones and laptops to enabling clean transportation and supporting grid-scale energy storage. Consequently, batteries have become indispensable in both daily life and strategic energy infrastructures. In particular, lithium-ion (Li^+) batteries, the dominant battery in the market, are facing increasingly stringent demands in terms of energy density, safety, recyclability, and mechanical flexibility[1–4], which have spurred the development of next-generation battery systems designed for specific applications. Considering the anticipated surge in retired batteries over the coming decade, battery recyclability is among the most pressing concerns to ensure environmental sustainability[5–7]. Simultaneously, the use of flammable organic liquid electrolytes in conventional Li^+ batteries has raised serious safety concerns. In this regard, solid-state batteries and aqueous metal-ion batteries have emerged as promising alternatives, with the former type effectively eliminating the risk of electrolyte leakage and the latter demonstrating inherent safety[8–12]. Moreover, the rapid advancement of flexible and wearable electronics has introduced new flexibility requirements for energy storage devices[13–15]. However, the rigid structure of current commercial batteries is incompatible with the flexibility and conformability needed for such applications[16]. The realization of next-generation battery systems heavily relies on material-level innovations, particularly the development of environmentally friendly and sustainable materials.

In this context, cellulose, the most abundant natural polymer on Earth, is a promising material offering versatile functionalization capacity, renewability, and degradability for the development of next-generation sustainable battery systems. Owing to their aligned, one-dimensional hierarchical structure and

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oxygen-containing polar functional groups, cellulose derivatives are highly attractive for high-performance solid-state batteries and aqueous metal-ion batteries. In addition, the robust mechanical properties and high aspect ratios of cellulose fibers render them suitable for the development of flexible electronics[17–20]. Moreover, electrochemical energy systems can benefit from the superior mechanical, optical, and functional properties of nanocellulose compared with those of bulky cellulose[21]. Given the growing interest in cellulose-based materials for sustainable energy applications, a systematic review is crucial to consolidate recent advances and guide future research directions.

This review systematically summarizes recent advances in the strategic integration of cellulose as a sustainable component into advanced energy storage systems. First, the fundamental characteristics and preparation strategies of cellulose are comprehensively described, including its tunable physicochemical properties and adaptive structure design, which align well with the stringent requirements of next-generation energy storage systems. Subsequently, the exploitation of these appealing characteristics in four critical energy storage components, i.e., electrode, solid electrolyte interphase (SEI), separator, and electrolyte, is discussed (Fig. 1). For instance, the abundant surface hydroxyl groups of cellulose confer exceptional water retention capacity, making it an ideal hydrogel electrolyte candidate for aqueous batteries. Finally, the review outlines future research directions, aiming at accelerating the development of sustainable cellulose-based energy storage solutions.

2 Why cellulose?

Cellulose is a linear homopolysaccharide composed of β-D-glucopyranose as a repeating unit linked by β-1,4-glycosidic bonds, with individual glucan chains exceeding 25,000 glucose residues. These macromolecules hierarchically assemble into highly packed flat ribbon structures via extensive intra- and intermolecular hydrogen bond networks; specifically, 15–45 glucan chains form paracrystalline microfibrils, which further aggregate into macrofibrils at the microscopic scale[22,23]. Most importantly, cellulose is a sustainable and abundant biomass resource that can be obtained via top-down and bottom-up strategies.

2.1 Top-down and bottom-up strategies to obtain cellulose

Lignocellulose, the primary constituent of agricultural and forestry residues, is globally the most abundant source of cellulose. Current land use patterns encompass approximately 3.2 billion acres of cropland, 8.4 billion acres of grazing land, and 10.1 billion acres of forests, annually generating millions of tons of lignocellulosic waste[24]. However, the predominant disposal methods—field accumulation and open-air incineration—not only waste this renewable resource but also increase greenhouse gas emissions[25]. Comprising 30 wt%–50 wt% cellulose, 20 wt%–35 wt% hemicellulose, and 15 wt%–30 wt% lignin, lignocellulose forms complex structural networks through covalent linkages between polysaccharides and aromatic lignin (Fig. 2a)[26,27]. The extraction of cellulose from lignocellulose represents a top-down strategy, for which various pretreatment techniques have been investigated. Interestingly, *in situ* functionalization can be simultaneously achieved during this process; for instance, carboxylated cellulose nanowhiskers were obtained via 2,2,6,6-tetramethylpipridine-1-oxyl (TEMPO) oxidation[36].

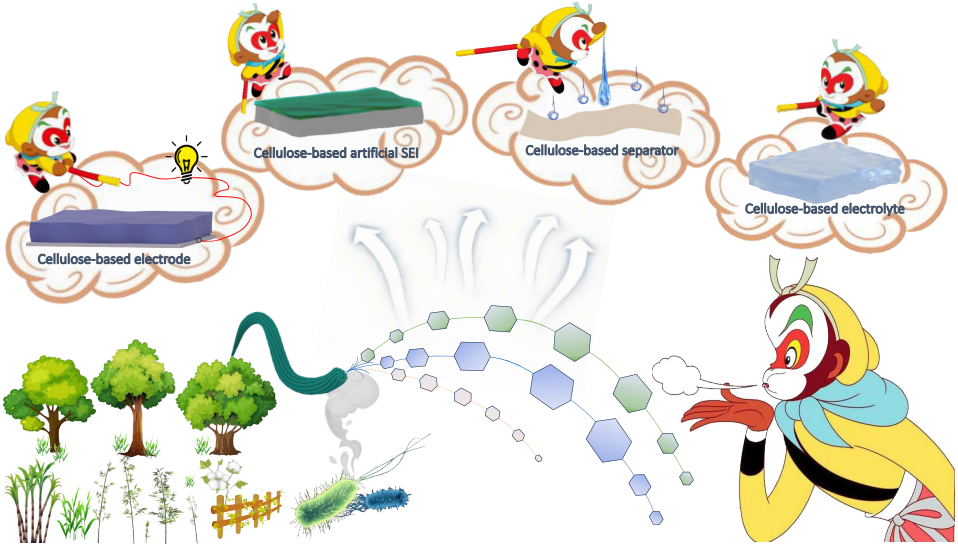

Figure 1 Schematic of cellulose applications in the electrode, solid electrolyte interface, separator, and electrolyte of energy storage devices.

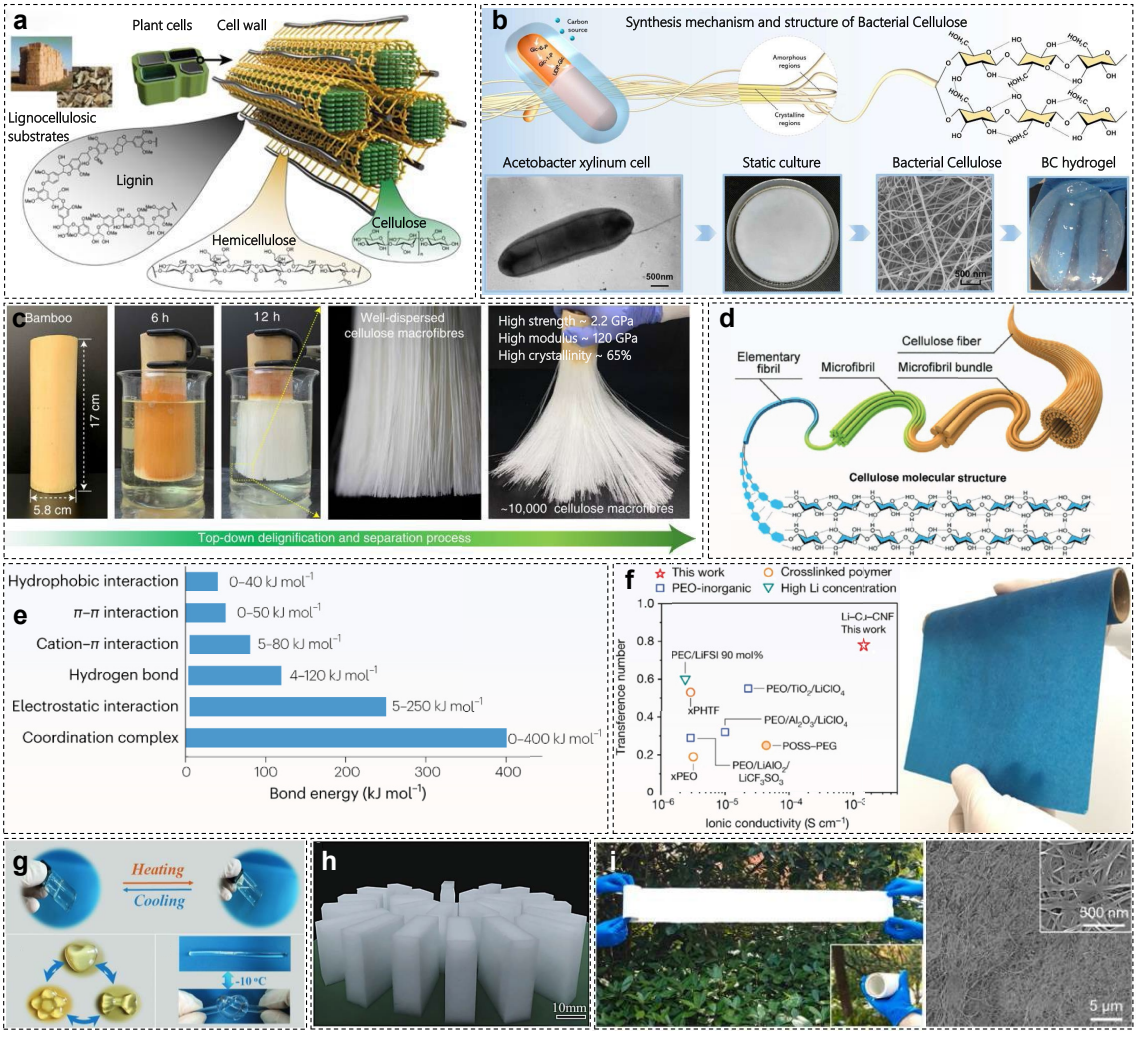

Figure 2 (a) Components and structure of lignocellulosic plant cell walls. Reproduced under the CC BY 4.0 license from Ref. [26] ©2020, The Authors. (b) Synthesis and hierarchical structure of bacterial cellulose. Reproduced with permission from Ref. [28] ©2023, Elsevier B.V. (c) Top-down scalable extraction of high-performance cellulose macrofibers from natural bamboo. Reproduced with permission from Ref. [29] ©2021, The Author(s), under exclusive licence to Springer Nature Limited. (d) Hierarchical structure and constituents of cellulose. Reproduced under the CC BY 4.0 license from Ref. [30] ©2025, The Authors. (e) Bond strengths of noncovalent interactions. Reproduced with permission from Ref. [31] ©2025, Springer Nature Limited. (f) Transference number vs. Li^+ conductivity and photograph of a Li–Cu–cellulose nanofibril membrane. Reproduced with permission from Ref. [32] ©2021, The Authors, under exclusive licence to Springer Nature Limited. (g) Photograph of a thermally reversible and antifreezing cellulose hydrogel. Reproduced with permission from Ref. [33] ©2019, Wiley-VCH GmbH & Co. KGaA, Weinheim. (h) Morphology of a lightweight insulating cellulose-based aerogel. Reproduced with permission from Ref. [34] ©2024, Wiley-VCH GmbH. (i) Morphology and structure of a sulfonated bacterial cellulose film. Reproduced with permission from Ref. [35] ©2025, Elsevier B.V.

Meanwhile, bacterial cellulose (BC) is synthesized by adopting a bottom-up strategy based on microbial fermentation (Fig. 2b)[28]. Structurally, BC consists of repeating D-glucose units covalently connected with β-1,4-glycosidic linkages, resembling the fundamental architecture of lignocellulose-derived cellulose. However, owing to the complete absence of hemicellulose and lignin in BC, its purity is superior to that of its lignocellulosic counterpart. Furthermore, the exceptional performance of BC stems from its distinctive nanofibrillar architecture, which features narrow fibers (10–100 nm in diameter) and remarkably high crystallinity (70%–90%)[37]. This structural characteristic is particularly advantageous for nonaqueous energy storage systems because the densely packed crystalline domains effectively inhibit water penetration into the microfibers. In addition, from a production perspective, BC holds considerable commercialization potential owing to the scalability and cost-effectiveness of microbial fermentation processes[38].

2.2 Mechanical and thermal properties

Using the aforementioned top-down and bottom-up preparation approaches along with diverse fabrication routes and treatments, cellulose-based materials can be endowed with numerous advantageous prop-

erties, among which their robust structural characteristics, particularly in nanocellulose, stand out. Nanocellulose exemplifies these distinctive structural characteristics, including aspect ratios of 200–5000, crystallinity degrees of 60%–80%, and specific surface areas approaching 500 $m^2 g^{-1}$[39]. This renewable material exhibits exceptional mechanical properties, particularly a tensile strength ranging from 2 to 7.7 GPa and an elastic modulus reaching approximately 150 GPa, while maintaining a remarkably low density of 1.6 g cm^{-3}[40]. Moreover, the thermal stability of nanocellulose further enhances its applicability, with thermal degradation occurring at elevated temperatures (200 °C–300 °C)[39]. These combined attributes position cellulose as a promising candidate for developing sustainable lightweight structural materials. For example, a scalable top-down strategy was developed for producing cellulose macrofibers from bamboo stems (Fig. 2c)[29]. The as-obtained macrofibers, which were characterized by densely packed, aligned cellulose nanofibers (CNF), demonstrated exceptional mechanical performance with a tensile strength of 1.90 GPa, a Young's modulus of 91.3 GPa, and a toughness reaching 25.4 MJ m^{-3} (Fig. 2c)[29]. Notably, the low density of this material contributes to a remarkable specific strength of 1.26 GPa cm^{-3} g^{-1}, outperforming traditional reinforcement materials[41]. Apart from one-dimensional high-aspect-ratio macrofibers, two-dimensional and mechanically robust monolithic lignocellulosic bioplastics were successfully fabricated using a biodegradable and scalable deep-eutectic-solvent strategy[42]. Owing to the entanglement of micro/nanofibrils and the integration of lignin-induced adhesion, the as-fabricated lignocellulosic bioplastic exhibited a remarkable tensile strength of ~128 MPa and a superior thermal degradation temperature of 357 °C, constituting a promising alternative to petroleum-based plastics.

2.3 Surface functionalization

In addition to its inherent biodegradability, exceptional mechanical strength, and remarkable thermal stability, cellulose exhibits superior functionalization capacity stemming from the presence of three hydroxyl groups (C2–OH, C3–OH, and C6–OH) per anhydroglucose unit (Fig. 2d)[30]. The reactivity of the hydroxyl groups in the cellulose skeleton displays position-dependent characteristics, with the primary C6–OH group showing the highest activity, followed by C2–OH and C3–OH[43]. The TEMPO-mediated oxidation at C6 position and phosphorylation at C2 and C6 positions exemplify this regioselectivity, which probably results from steric hindrance[44,45]. The presence of multiple reactive hydroxyl groups in cellulose provides versatile chemical modification platforms, enabling the synthesis of diverse functional derivatives through well-established reaction pathways, including sulfonation[46], esterification[47], acetylation[48], and oxidation processes[49]. For instance, uniform esterified cellulose films with smooth surfaces were obtained via surface esterification with short-chain carboxylic acids and scalable spray coating[50]. The esterification considerably increased the tensile strength by ~30 MPa, demonstrating that esterified cellulose films are viable alternatives to traditional petroleum-based plastics. Moreover, surface acetylation was employed to regulate the surface hydrophobicity of cellulosic materials, affording acetylated cellulose, which exhibited great potential as a substitute for the cellulose acetate used in films, textiles, and plastics[48]. When integrated in energy storage devices, these functionalization designs could suppress dendrite growth and hydrogen evolution reactions, thus contributing to enhancing the electrochemical performance[51,52].

2.4 Engineering cellulose nanocomposites via supramolecular chemistry

Supramolecular chemistry focuses on noncovalent molecular interactions such as hydrogen bonding, electrostatic forces, metal coordination, host–guest interactions, and hydrophobic effects (Fig. 2e). These interactions are typically weak, reversible, and highly tunable, covering a broad energy range from ~40 kJ mol^{-1} for hydrophobic interactions to ~400 kJ mol^{-1} for strong metal coordination complexes. This tunability offers a powerful approach for engineering cellulose nanocomposites with targeted mechanical, chemical, and biological properties[31]. For example, metal coordination of Cu^{2+} ions can expand the molecular channels within CNF, enabling a rapid Li^+ transport along the polymer chains of otherwise ion-insulating cellulose (Fig. 2f)[32]. The resulting Cu^{2+}-coordinated cellulose ion conductors exhibited advantageous characteristics, including high Li^+ conductivity (1.5 mS cm^{-2}), a high transference number (0.78), and a wide electrochemical stability window (0–4.5 V) compatible with common battery electrodes. Host–guest interactions represent another key strategy; incorporating such systems into cellulose matrices can endow them with self-healing and dynamic properties. For instance, crown ether derivatives featuring electron-donating cyclic cavities can be integrated into cellulose structures to act as carriers, facilitating cation transport in polymer membranes[53].

2.5 Adaptable monolithic structures

Cellulose fibrils can be controllably restructured into precisely organized hydrogel networks[33], hierarchically porous aerogels[34], and freestanding thin films[35] using various well-established strategies. For example, Zhang et al. prepared cellulose hydrogels exhibit-

ing thermal reversibility and freeze resistance while maintaining ion conductivity (Fig. 2g)[33]. These materials showed critical mechanical compliance and electrical conductivity even under ultralow temperature conditions, enabling their application in flexible devices designed for extreme environments. In addition, a scalable colloidal self-assembly strategy utilizing Earth-abundant wood biopolymers enabled the fabrication of nanostructured silanized cellulose frameworks with multidimensional gradient pore architectures (Fig. 2h)[34]. The engineered three-dimensional nanoporous aerogels exhibited homogeneous surfaces with exceptional specific surface area (875.8 $m^2 g^{-1}$) and ultrahigh porosity (99.7%). Moreover, the resulting lightweight silanized cellulose aerogels showed an ultralow thermal conductivity of 15.9 mW $m^{-1} K^{-1}$, which is beneficial for practical thermal insulation applications. Recently, scalable and sustainable sulfonated BC films exhibiting a minimal thickness of 50 μm, a substantial wet tensile strength of 167 MPa, and a remarkable ionic conductivity of 13.1 mS cm^{-1} were developed (Fig. 2i)[35]. Critically, the incorporated sulfonate groups imparted exceptional ion-sieving functionality, rendering the obtained cellulose films suitable as separators in energy storage devices.

In summary, cellulose stands out as a multifunctional biopolymer that uniquely integrates renewability, biodegradability, mechanical resilience, thermal stability, and tunable chemistry. Moreover, its unique three-dimensional structure enables the production of precisely organized hydrogels, multilevel porous aerogels, and freestanding thin films. These cellulose-derived monolithic structures exhibit potential applicability in the critical components of energy storage systems: electrode, SEI, separator, and electrolyte. The following sections provide an overview of recent advances in such applications.

3 Cellulose in the electrode

The polar functional groups within cellulose facilitate interfacial interactions such as hydrogen bonding, ion–dipole interactions, and chemical bonding[54]. Furthermore, owing to its intrinsic porosity and flexibility, the incorporation of cellulose into a substrate or structured network promotes the ion transport, enhancing the electrochemical performance[55]. These characteristics make cellulose particularly relevant for electrode applications, where binders are essential components. Binders provide adhesion between active material particles and current collectors, ensure interparticle cohesion, deliver the slurry rheology required for processing, and maintain mechanical integrity during densification and cycling[56]. Consequently, an optimal binder must enable a homogeneous slurry formation, exhibit balanced adhesiveness and electrochemical stability, facilitate ion transport, and be cost-effective and sustainable[57]. Cellulose micro/nanofibers combine these multidimensional requirements, constituting a promising building block while preserving the microstructure of electrode materials (Fig. 3a)[58].

3.1 Cellulose-based binders

Current industrial binders such as polyvinylidene fluoride (PVDF) require dissolution in toxic and expensive solvents like *N*-methylpyrrolidone. Furthermore, PVDF primarily interacts with electrode materials through weak H–F van der Waals forces, providing insufficient structural support[54]. By contrast, cellulose fibers serve as effective binders and dispersing agents in composite electrodes. Their polar functional groups (hydroxyl, carboxyl, and ether groups) enable strong interfacial interactions with active materials, including hydrogen bonding, ion–dipole interactions, and chemical bonding[54]. For example, a molecular ligand-mediated layer-by-layer assembly strategy was proposed to bridge all the interfaces of conductive metal and metal oxide nanoparticles[59], affording nanocomposite papers demonstrating an exceptionally low sheet resistance of 0.31 Ω sq^{-1} and a high electrical conductivity of 230.1 S cm^{-1} without requiring additional thermal or mechanical treatment (Fig. 3b). The resulting porous metallic paper functioned simultaneously as both current collector and reservoir for high-energy pseudocapacitive nanoparticles. Consequently, supercapacitors incorporating this metallic paper exhibited maximum power and energy densities of 15.1 mW cm^{-2} and 267.3 μWh cm^{-2}, respectively, outperforming conventional paper or textile-based supercapacitors. Moreover, in MXene/cellulose composites, cellulose can act as a network structure substrate to enhance the flexibility and processability of MXene and expand its application areas through an appropriate structural design[63]. For instance, CNF were introduced as binders and intercalating agents to fabricate three-dimensional macroporous MXene/CNF/reduced graphene oxide aerogel electrodes (Fig. 3c)[60], resulting in an exceptional electrochemical performance with an areal specific capacitance of 671 mF cm^{-2}, an energy density of 60.9 mWh cm^{-2}, and 79.4% retention rate after 5000 cycles at a power density of 1.0 mW cm^{-2}.

3.2 Cellulose-based thick electrodes

Enhancing the energy storage density predominantly relies on thick electrodes with a high active material loading, which are traditionally produced using coating methods. However, these electrodes face practical limitations in high-energy applications owing to restricted ion transport, insufficient mechanical stability, and reduced energy density stemming from an excessive current collector mass. In this context,

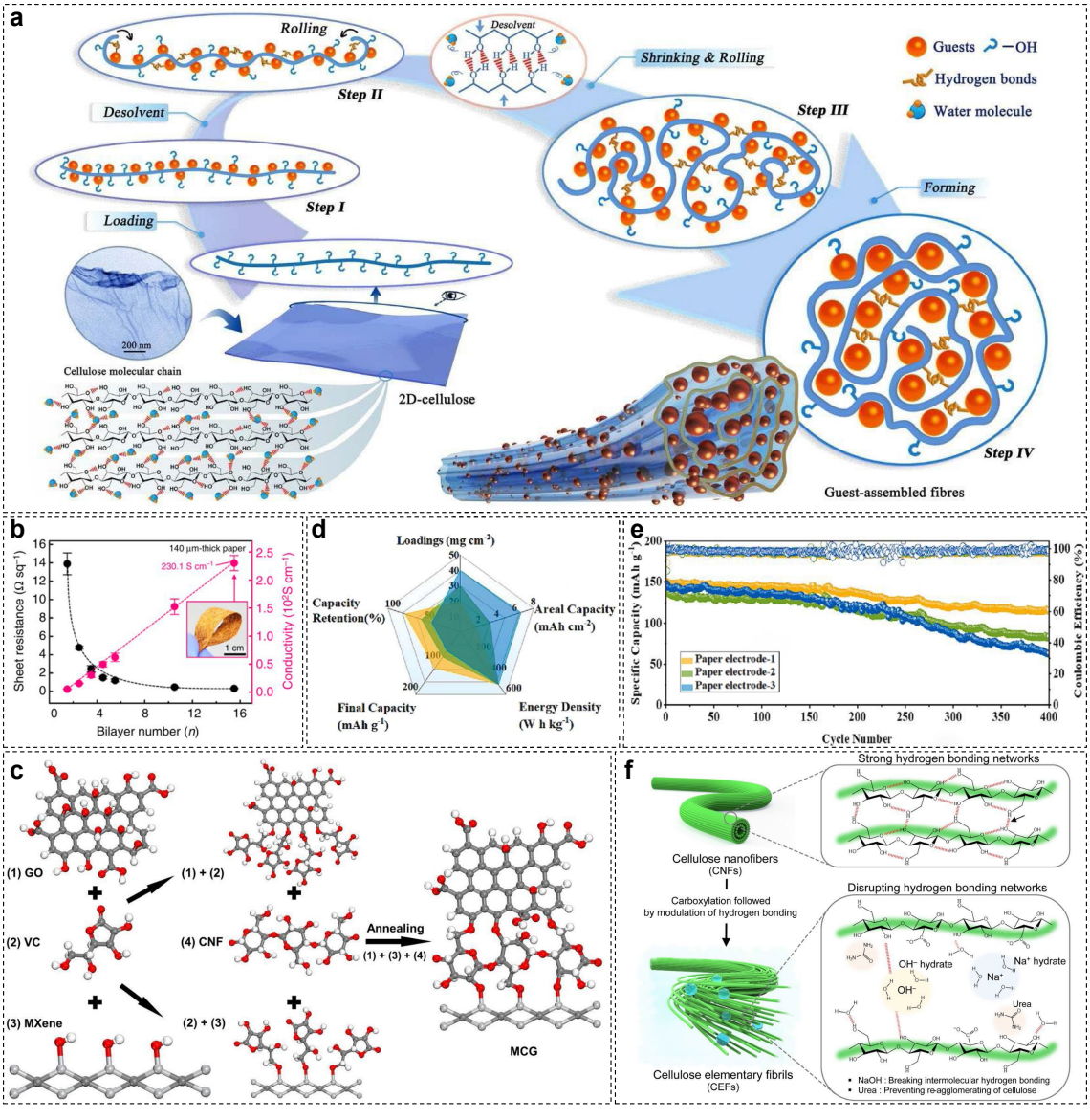

Figure 3 (a) Conversion mechanism of powders into micro/nanofibers mediated by a two-dimensional cellulose isomer during water removal. Reproduced with permission from Ref. [58] ©2024, The Authors, under exclusive licence to Springer Nature Limited. (b) Electrical properties of metallic paper with increasing bilayer number (*n*). Reproduced under the CC BY 4.0 license from Ref. [59] ©2017, The Authors. (c) Self-assembly between graphene nanosheets and $Ti_3C_2T_x$ MXene via the bridge-linking effect of CNF. GO, graphene oxide; VC, L-ascorbic acid; MCG, MXene/CNF/reduced graphene oxide. Reproduced with permission from Ref. [60] ©2024, The Authors, under exclusive licence to Springer Nature Switzerland AG. (d) Performance hexagons of paper electrodes with different loading amounts. (e) Cycle performance of paper electrodes with different loading amounts. Reproduced with permission from Ref. [61] ©2024, Published by Elsevier B.V. (f) Schematic of the hierarchical structure and fibrillation mechanism of cellulose. Reproduced under the CC BY 4.0 license from Ref. [62] ©2025, The Authors.

cellulose materials serve as promising binders that mitigate particle agglomeration in high-mass-loading electrodes, leveraging their inherent porosity to facilitate ion diffusion and enhance electrolyte contact with active sites[61,62]. For example, Jiang et al. developed a continuous three-dimensional porous conductive network via the self-assembly of carbon nanoparticles onto negatively charged natural cellulose fibers[61]. The resulting integrated structure substantially improved both ion and electron transport pathways, enabling a high active material loading of 40 mg cm^{-2} and exceptional mechanical integrity in a binder-free electrode architecture (Fig. 3d). Owing to the robust electrode structure and fast transfer kinetics of electrons and Li^+, the batteries prepared using these electrodes exhibited excellent cycle stability with a capacity retention of 98% after 400 cycles, thereby demonstrating the effectiveness of cellulose in advanced electrode design (Fig. 3e). Moreover, to regulate the intermolecular hydrogen bonding in CNF, a treatment employing NaOH as a proton acceptor and urea as a hydrotropic agent was applied (Fig. 3f)[62]. This process facilitated the deagglomeration of CNF aggregates into individualized

cellulose elementary fibrils, which were subsequently utilized as a deagglomerated binder in the fabrication of high-mass-loading electrodes. The corresponding cathode achieved notably high areal mass loading (50 mg cm^{-2}, equivalent to an areal capacity of 12.5 mAh cm^{-2}) and specific energy density (445.4 Wh kg^{-1}), highlighting the supporting role of cellulose-derived binders in sustainable high-mass-loading electrode architectures.

4 Cellulose in SEI

The inherent mechanical robustness of cellulose enables the fabrication of freestanding nanoscale membranes featuring aligned molecular nanochannels, which can spatially regulate ion deposition and improve electrochemical reversibility. Furthermore, its capacity for functionalization allows grafting diverse functional groups, enhancing the interfacial kinetics at the electrode interface. This interface is critically governed by the SEI, a layer physically separating the electrolyte and the electrode. The main function of the SEI is to electronically insulate the electrode while facilitating ion transport; therefore, its quality directly influences the charge transfer kinetics and ultimately determines the overall electrochemical performance. Consequently, cellulose-based materials, which inherently provide electronic insulation and allow regulating ion transport through their modification, represent an ideal class of materials for constructing artificial SEI layers.

4.1 Cellulose-based nanoscale membranes

Moving beyond conventional artificial SEI layers as homogeneous nanoscale membranes, cellulose-derived ultrathin membranes are now positioned as viable alternatives[64]. By leveraging their inherent structural advantages, including mechanical resilience, abundant polar functional groups, and ion transport modulation capabilities, these bio-based layers exhibit enhanced electrochemical reversibility during repeated cycling. For instance, Lu et al. developed a biomimetic SEI layer via top-down exfoliation of 10 nm-thick pit-membrane-like nanowood (Fig. 4a)[65]. This engineered interface features naturally aligned cellulose molecular nanochannels that spatially regulate Li^+ deposition, coupled with interconnected micropores and lithiophilic groups that accelerate Li^+ transport. The binding energies of carboxyl (E_{b1}, 15.81 kcal mol^{-1}), hydroxyl (E_{b2}, 7.35 kcal mol^{-1}), phenolic hydroxyl (E_{b3}, 7.15 kcal mol^{-1}), and methoxyl groups (E_{b4}, 6.85 kcal mol^{-1}) are higher than that of a commercial polypropylene separator (6.61 kcal mol^{-1}), indicating that Li^+ interacts more strongly with these four species (Figs. 4b and 4c). This synergistic architecture endows Li^+ batteries with a regulated ion flux, achieving a remarkable capacity of ~140 mAh g^{-1} and an average Coulombic efficiency of 99.6% over 800 cycles. Moreover, Lu et al. developed hydroxyethyl cellulose (HEC) membranes as efficient artificial SEI layers for zinc (Zn) metal electrodes (Fig. 4d)[66]. Combined experimental and computational analyses revealed that the hydroxyl and ether groups in HEC selectively interact with hydrated zinc ions (Zn^{2+}), inducing the removal of water from the solvation sheath of Zn^{2+}. The binding energy of hydrated Zn^{2+} is −1297.26 kJ mol^{-1}, whereas that of the HEC–Zn^{2+} complex is −1414.88 kJ mol^{-1} (Fig. 4e). This lower binding energy indicates that the HEC–Zn^{2+} complex is thermodynamically more stable. Consequently, this mechanism simultaneously suppresses hydrogen evolution and inhibits dendrite growth from zinc hydroxide sulfate by blocking water contact at the electrode surface. The engineered HEC-coated Zn electrodes demonstrated exceptional cyclability in symmetric cells, maintaining a stable operation for 2,700 h at 5 mA cm^{-2}/5 mAh cm^{-2}. In addition, amino-functionalized BC membranes with fast ion transport kinetics were engineered as artificial SEI layers to address Zn anode challenges (Fig. 4f)[67]. The chelation between the amino groups and Zn^{2+} promoted Zn^{2+} deposition along the (002) crystallographic plane while preventing dendrite growth, thereby concurrently mitigating the reactivity of hydrated Zn^{2+} and water-induced side reactions.

4.2 Cellulose-based *in situ* SEIs

Cellulose-based materials have garnered considerable attention for *in situ* SEI fabrication[69,68]. This approach represents a particularly promising strategy owing to its procedural simplicity and cost-effectiveness. For instance, the *in situ* protective layer strategy employing bio-derived cellulose levulinate ester (CLE) additives with acetyl propyl ketone moieties effectively regulates the Zn anode–electrolyte interfacial chemistry (Fig. 4g)[68]. The synergistic interactions among ether bonds, hydroxyl and ester groups, and keto–enol tautomerism accelerate Zn^{2+} desolvation while suppressing dendrite growth and parasitic reactions. The $Zn(OTf)_2$/CLE electrolyte enables a stable Zn^{2+} plating/stripping for over 2,800 h at 1 mA cm^{-2} in shelving-recovery tests, providing good electrochemical performance. In fact, when integrated with MnO_2 cathodes in full cells, the system demonstrated 78.6% capacity retention after 3,000 cycles at 2 A g^{-1}, confirming the efficiency of the *in situ* protective layer in practical aqueous Zn^{2+} batteries.

5 Cellulose in the separator

Extensive inter- and intramolecular hydrogen bond-

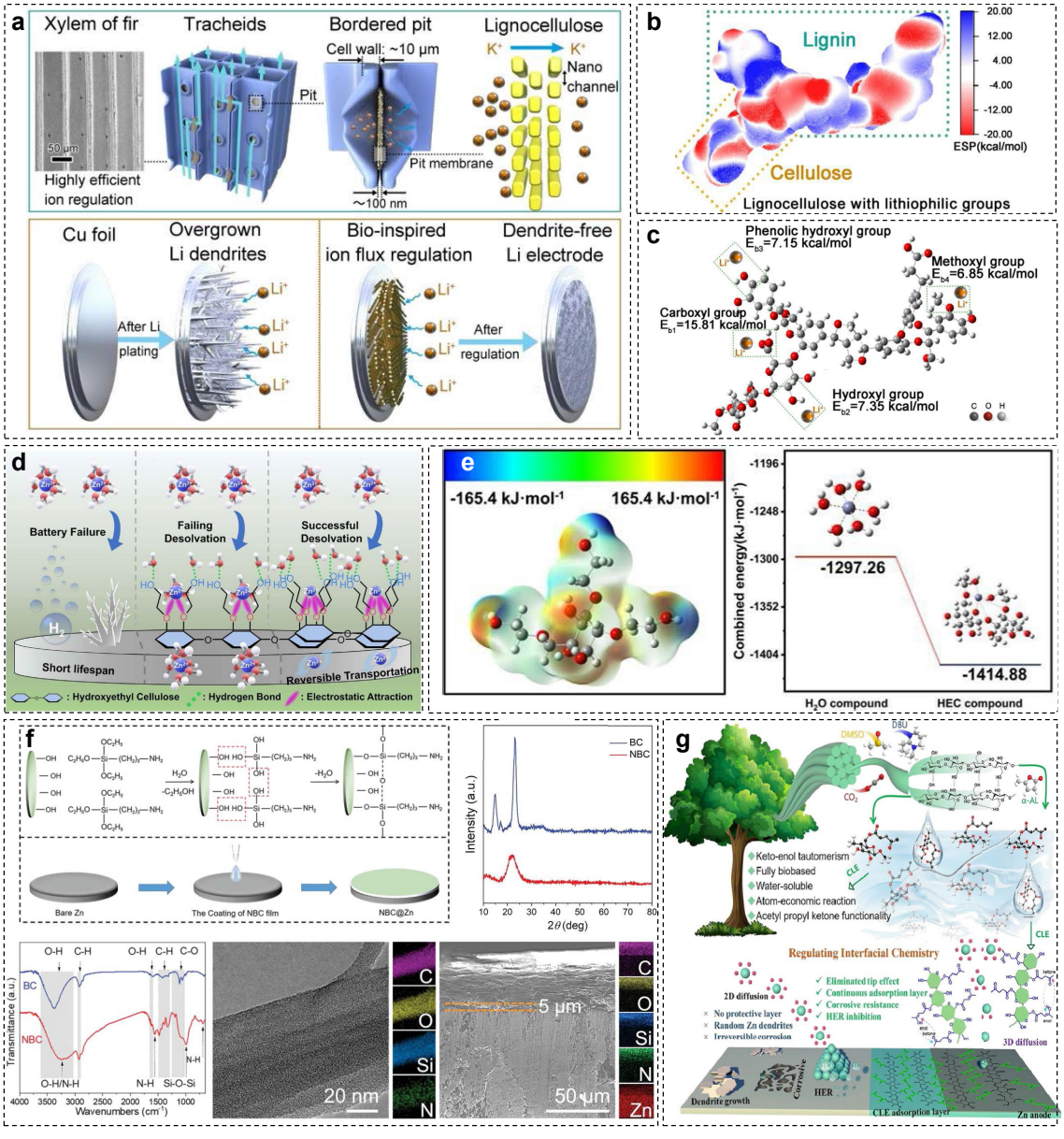

Figure 4 (a) Schematic illustration of the pit-membrane design and construction of the wood-structure-based artificial solid electrolyte interphase for Li metal anodes. Molecular electrostatic potential (ESP) (b) and optimized structure (c) showing the strong interactions between Li^+ and the polar functional groups of lignocellulose within the nanowood film. Reproduced with permission from Ref. [65] ©2021, American Chemical Society. (d) Schematic of the transport mechanism of Zn^{2+} and $Zn(H_2O)_6^{2+}$ across the bare thin Zn surface and optimal HEC layers in aqueous Zn^{2+} batteries. (e) Electrostatic potential distribution of HEC and binding energies of Zn^{2+} with water and HEC. Reproduced with permission from Ref. [66] ©2024, The Royal Society of Chemistry. (f) Ultrauniform solid electrolyte interphase from an amino-functionalized interfacial layer for high-performance aqueous Zn^{2+} batteries. Reproduced with permission from Ref. [67] ©2023, Wiley-VCH GmbH. (g) Regulating interfacial chemistry with bio-based multifunctional CLE for highly reversible Zn^{2+} batteries. Reproduced with permission from Ref. [68] ©2024, Elsevier B.V.

ing endows cellulose with an excellent film-forming ability, making it well-suited for separator applications in energy storage systems. The separator serves as an insulating porous barrier between electrodes, preventing internal short circuits while providing ion transport channels[70]. Consequently, an optimal separator must exhibit cost-effectiveness, superior electrolyte wettability, high thermal and electrochemical stability, moderate porosity with uniform pore distribution, sufficient thickness to maintain integrity in the wet state, and adequate mechanical strength for safe operation[71–75]. In this context, cellulose emerges as a highly promising separator material fulfilling these critical requirements owing to its exceptional mechanical strength, excellent hydrophilicity, versatile functionalization capacity, and effective insulating properties.

5.1 Cellulose-based separators

To ensure battery safety, separators must exhibit

substantial tensile strength while resisting elongation under tension to prevent dimensional changes[72,76]. Meanwhile, the separator electrochemical performance is primarily governed by the ion transport efficiency, which depends critically on electrolyte wettability and pore uniformity. By virtue of its superior mechanical robustness stemming from its covalent framework and extensive inter/intramolecular hydrogen bonding, inherent porosity, and polar functional groups, cellulose meets these stringent requirements[77]. For example, Huang et al. fabricated an ultrathin regenerated cellulose separator (16 μm) exhibiting impressive mechanical properties with a tensile strength of 32.6 MPa and an elongation at break of 8.4% (Figs. 5a and 5b)[74]. In addition, the high thermal stability (200 °C), high porosity (65.4%) with a uniform pore structure, and exceptional electrolyte adsorption capacity (82%) contributed to enhancing the ion transport performance (9.2 mS cm^{-1}) (Figs. 5c and 5d). Zhou et al. introduced a sulfonated cellulose separator featuring an optimized combination of ultrathin profile (~50 μm), superior wet mechanical resilience (18.7 MPa), and enhanced ionic conductivity (52.1 mS cm^{-1})[46]. The engineered sulfonic groups demonstrated simultaneous dual functionality: blockage of sulfate migration

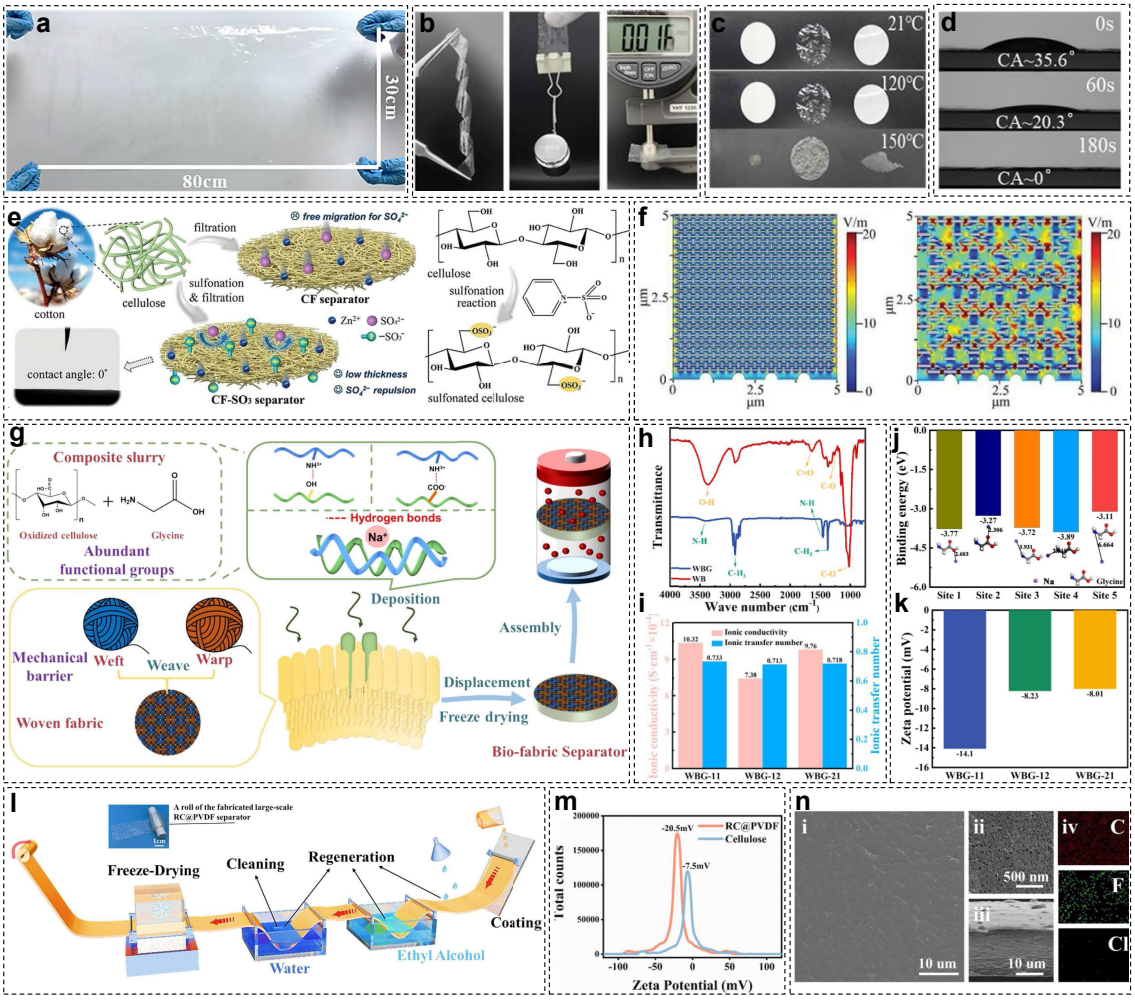

Figure 5 (a) and (b) Photographs of a regenerated cellulose separator. (c) Photographs showing the thermal shrinkage of different separators (left: commercial non-woven polypropylene separator; middle: regenerated cellulose; right: commercial polypropylene separator). (d) Pictures of the contact angle (CA) of a regenerated cellulose separator at different times. Reproduced with permission from Ref. [74] ©2024, The Royal Society of Chemistry. (e) Schematic of the preparation process of a sulfonated cellulose separator. CF, cellulose fibers. (f) Electric field at the surface of a Zn electrode with a sulfonated cellulose separator (left) and with a glass fiber separator (right). Reproduced with permission from Ref. [46] ©2024, Wiley-VCH GmbH. (g) Schematic of the preparation of a biofabric separator. (h) FT-IR patterns of biofabric separators. WBG, woven fabric-based TEMPO-oxidized bacterial cellulose–glycine composite separator; WB, woven fabric separator coated with oxidized bacterial cellulose. (i) Ionic conductivity and ionic transport number of biofabric separators. (j) Na binding energies with glycine in the biofabric separator. (k) Zeta potential of the biofabric separator. Reproduced with permission from Ref. [78] ©2024, Elsevier Ltd. (l) Phase transformation strategy to fabricate a composite separator made of porous regenerated cellulose and polyvinylidene difluoride (RC@PVDF). (m) Zeta potential of the RC@PVDF separator and a cellulose separator. (n) SEM images and elemental analysis of a regenerated cellulose separator. Reproduced with permission from Ref. [79] ©2024, Elsevier B.V.

via electrostatic repulsion and increase in Zn^{2+} transfer (Fig. 5e). These sulfonic groups establish strong coordination interactions with Zn^{2+}, promoting the desolvation of hydrated Zn^{2+} and suppressing their planar diffusion across the electrode surface. Finite element simulations also demonstrated that the sulfonated cellulose separator achieved a much more uniform distribution of the electric field compared with a glass fiber separator (Fig. 5f). Similarly, a TEMPO-mediated oxidation strategy was employed to fabricate an anionic carboxylate-functionalized CNF separator[80]. The negatively charged carboxylate groups also exhibited ion-sieving function, preventing the shuttling of sulfate via electrostatic repulsion while boosting the Zn^{2+} transfer, which was beneficial for maintaining the stability of the Zn anode without dendrite formation and adverse reactions.

5.2 Cellulose-based composite separators

The fabrication of composite separators enables the synergistic integration of the beneficial properties of the individual components. For example, Zhang et al. fabricated a bio-derived composite separator via sequential deposition of oxidized BC and glycine onto a polypropylene woven substrate to address limitations in ionic regulation (Fig. 5g)[78]. The resulting composite structure incorporated electronegative functional groups (carboxyl, amino, and hydroxyl groups) with high sodium (Na) affinity, thereby enabling controlled sodium-ion (Na^+) migration and uniform deposition (Figs. 5h and 5i). The stable adsorption configurations of glycine at five different Na adsorption sites indicated that glycine has good affinity for Na^+, which facilitated Na^+ transfer (Fig. 5j). Further, zeta potential tests on biofabric separators with three different ratios demonstrated that all biofabric separators exhibited electronegativity, which contributes to the rapid migration of positively charged Na^+ (Fig. 5k). This biomolecular synergy substantially enhances the Na^+ transport characteristics while concurrently improving the mechanical robustness. Similarly, Huang et al. engineered a composite separator (RC@PVDF) by integrating the hierarchical porosity of regenerated cellulose with the electronegativity of PVDF (Fig. 5l)[79]. This design leveraged aligned cellulose nanofibers to create optimized ion channels while utilizing fluorinated domains for electrostatic selectivity. The zeta potential of the RC@PVDF separator surface was −20.5 mV, proving its strong electronegativity in the absence of an electric field (Fig. 5m). Energy-dispersive spectroscopy elemental mapping indicated that electronegative fluorine atoms were successfully incorporated into the RC@PVDF separator (Fig. 5n). Moreover, the intermolecular hydrogen bonding between the components provided enhanced thermal stability and mechanical strength, collectively demonstrating the advantages of the synergistic material combination.

6 Cellulose in the electrolyte

The inherent processability and three-dimensional network-forming ability of cellulose enable the fabrication of bio-based solid and quasi-solid polymer electrolytes. Because the electrolyte composition critically determines the battery performance, particularly for high-energy-density and functional systems, advanced formulations must fulfill the following requirements: high ionic conductivity for an efficient charge transfer coupled with low electronic conductivity to prevent self-discharge, a suitable electrochemical stability window to resist decomposition during cycling, excellent electrode compatibility to enhance the energy density, thermal stability, cost-effectiveness, manufacturing feasibility, and low toxicity, especially for grid-scale applications[81,82]. Cellulose-derived solid and quasi-solid electrolytes inherently address these multifaceted demands, leveraging the superior mechanical strength for structural integrity, the three-dimensional network for an efficient ion transport, and the versatile functionalization capacity for a uniform ion deposition.

6.1 Cellulose-based solid-state electrolytes

Cellulose-based materials have garnered much attention as solid-state electrolytes owing to their exceptional processability, abundant hydroxyl groups, and three-dimensional network-forming ability. For example, Li et al. demonstrated an ecofriendly manufacturing process converting cellulose into cellulose phthalate via esterification with phthalic anhydride (Fig. 6a)[83]. Mechanistic analyses verified the dual effects of phthalate modification: facilitating multiple oxygen interactions with Li^+ to establish efficient conduction pathways and reinforcing hydrogen bond networks to maintain mechanical stability. The practical implementation of solid-state electrolytes is often hindered by limitations including low ionic conductivity and high brittleness, particularly in inorganic ceramic-type electrolytes. To address these challenges, Yang et al. developed a three-dimensional solid polymer electrolyte by integrating rigid cellulose nanocrystals as a reinforcing framework with flexible polyacrylonitrile as an ion-conducting matrix[88]. The composite system demonstrated exceptional processability requiring an ultralow amount of solvent, achieving lightweight characteristics (1.2 g cm^{-3}) while maintaining structural integrity under operational stress. Moreover, Xie et al. developed a BC-templated hybrid electrolyte system combining $Li_7La_3Zr_2O_{12}$ with poly(ethylene oxide) (PEO) (Fig. 6b)[84]. The engineered well-organized network established continuous ion conduction pathways while increasing free Li^+ concentration

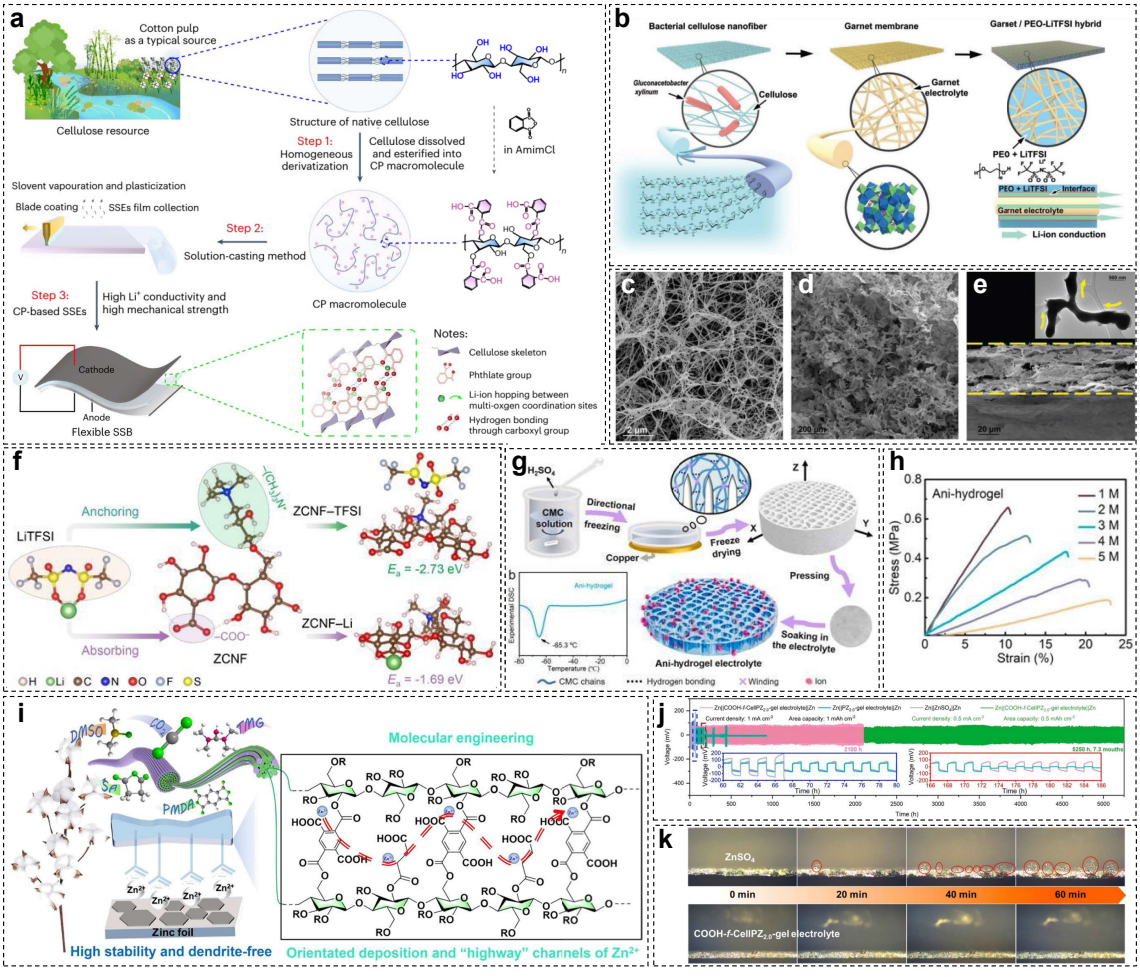

Figure 6 (a) Schematic of the fabrication of cellulose phthalate-based solid-state electrolytes. Reproduced with permission from Ref. [83] ©2024, The Authors, under exclusive licence to Springer Nature Limited. (b) Synthesis of a hybrid electrolyte using bacterial cellulose as an inexpensive, scalable, and efficient template. (c)–(e) SEM images of the cellulose fibrils, the $Li_7La_3Zr_2O_{12}$ membrane after calcination, and a cross-section view of the $Li_7La_3Zr_2O_{12}$ nanofiber network. Reproduced with permission from Ref. [84] ©2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (f) Mechanism of the interactions between zwitterionic cellulose nanofibrils (ZCNF) and lithium bis(trifluoromethanesulfonyl) imide (LiTFSI). Reproduced with permission from Ref. [85] ©2024, Wiley-VCH GmbH. (g) Anisotropic and antifreezing cellulose hydrogel electrolyte with aligned channels stabilizing a Zn metal anode. (h) Tensile stress–strain curves of hydrogel electrolytes using different concentrations of $Zn(ClO_4)_2$. Reproduced with permission from Ref. [86] ©2025, Elsevier B.V. (i) Molecularly engineered carboxylic acid-functionalized cellulose hydrogel electrolyte for highly stable Zn^{2+} hybrid capacitors. (j) Voltage–time profiles of Zn||Zn cells using different electrolytes. (k) *In situ* optical microscopy observations of Zn electrodeposition with a cellulose-based hydrogel electrolyte. Reproduced with permission from Ref. [87] ©2023, Elsevier B.V.

(Figs. 6c–6e). Notably, the hybrid electrolyte presented structural flexibility with a minor impedance increase after bending, overcoming the brittleness of the inorganic electrolyte. Similarly, to address the inferior ionic conductivity and insufficient mechanical robustness of PEO-based solid-state electrolytes, Cheng et al. engineered a PEO-based composite electrolyte reinforced with zwitterionic cellulose nanofibrils (ZCNF)[85]. The interactions between ZCNF and lithium bis(trifluoromethanesulfonyl) imide (LiTFSI) revealed that ZCNF capture and interact with $TFSI^-$ (Fig. 6f). Further, the zwitterionic nature of ZCNF concurrently disrupted PEO crystallization, enhanced Li salt dissociation, and accelerated Li^+ migration via electrostatic interactions, while inherently providing structural reinforcement.

6.2 Cellulose-based quasi-solid-state gel-based electrolytes

Quasi-solid-state hydrogel electrolytes offer superior ionic conductivity and interfacial compatibility compared with their solid-state counterparts[89,90]. Cellulose derivatives serve as promising structural frameworks for these hydrogels because their inherent three-dimensional hierarchical architecture provides mechanical reinforcement while facilitating ion transport[91]. This capability was exploited by Zhang et al., who fabricated an anisotropic carboxymethyl cellulose hydrogel electrolyte via directional freezing to create aligned ion transport channels that mitigated Zn dendrite growth and

parasitic reactions in aqueous Zn^{2+} batteries (Fig. 6g)[86]. The engineered hydrogel achieved an ultralow freezing point of −65.3 °C while exhibiting high tensile strength (0.45 MPa) and improved ionic conductivity (27.35 mS cm^{-1}) (Fig. 6h). Similarly, Chen et al. developed a carboxylic acid-functionalized cellulose hydrogel electrolyte using the unique solution properties of cellulose in superbase/DMSO/CO_2 systems[87]. This *in situ* derivatization strategy simultaneously disrupted native hydrogen bond networks and introduced carboxyl groups, creating unobstructed ion transport channels while modulating the Zn deposition morphology (Fig. 6i). Consequently, the optimized cellulose-based hydrogel electrolytes showed exceptional long-term cycling stability in Zn symmetric cells (over 5250 h at 0.5 mA cm^{-2}/0.5 mAh cm^{-2}), demonstrating that cellulose meets the critical requirements of hydrogel electrolytes (Figs. 6j and 6k).

7 Remaining challenges

Despite the notable progress made in cellulose-based rechargeable batteries, as outlined in Table 1, several key challenges remain to be addressed to realize their broad application.

First, the inherent hygroscopicity of cellulose fibers contradicts the stringent moisture control required in Li^+ and Na^+ battery systems. For instance, in Li^+ batteries, the water content must be lower than 20–50 ppm to prevent salt degradation; however, cellulose fibers can retain moisture levels ranging from 2×10^4 to 12×10^4 ppm at room temperature[92]. Nevertheless, prolonged thermal treatment at temperatures under 200 °C effectively removes the majority of adsorbed water from cellulose, thereby mitigating this limitation.

Second, cellulose is generally regarded as a poor thermal conductor owing to its molecular and crystalline structure, although the conductivity improves with increasing crystallinity[93]. This is a major issue because an efficient thermal management is crucial in electronics to rapidly dissipate heat, preventing heat accumulation that can impair device reliability or lead to safety hazards. Integrating cellulose with highly conductive materials such as boron nitride or graphene offers a promising route to enhance the thermal conductivity[94].

Third, cellulose exhibits limited ionic conductivity, primarily because of its high glass transition temperature and poor solvation ability for metal salts, often resulting in conductivities below the practical threshold[31]. Introducing specific metal ions such as Cu^{2+} can enlarge the spacing between cellulose chains, facilitating ion transport[32].

Finally, scalability remains a major obstacle for commercial applications. Moving from lab-scale to industrial production requires addressing issues such as sustainable sourcing, cost-effective manufacturing, and consistent quality. Addressing these challenges will pave the way for the widespread use of low-cost, high-energy-density cellulose-based electrochemical devices.

8 Summary and outlook

Cellulose exhibits great promise for application in energy storage devices owing to its multifaceted char-

Table 1 Summary of cellulose integration into energy storage devices.

Cellulose	Anode	Separator	Electrolyte	Cathode	Capacity/current density	Reference
CMC	CMC/LS-integrated HC	glass fiber	$NaPF_6$	Na sheet	348.0 mA h g^{-1} (0.05 A g^{-1})	[54]
CMC	Li metal	NM	$LiPF_6$	CMC-integrated $LiFePO_4$	165.9 mAh g^{-1} (0.1 C)	[61]
CEF	Li metal	NM	$LiPF_6$	CEF-based OLO	250.0 mAh g^{-1} (0.1 C)	[62]
TOCNF	Zn@TOCNF	nonwoven fiber film	$ZnSO_4/MnSO_4$	MnO_2	100.0 mAh g^{-1} (5 A g^{-1})	[64]
Nanowood	Li@Nanowood	Celgard 2400	LiTFSI	$LiFePO_4$	140.0 mAh g^{-1} (0.5 C)	[65]
HEC	Zn@HEC	glass fiber	$ZnSO_4/MnSO_4$	MnO_2	~70.0 mAh g^{-1} (10 C)	[66]
NBC	Zn@NBC	glass fiber	$ZnSO_4$	V_2O_5	201.0 mA h g^{-1} (0.5 A g^{-1})	[67]
CLE	Zn metal	glass fiber	$Zn(OTf)_2$/CLE	MnO_2	260.7 mA h g^{-1} (0.2 A g^{-1})	[68]
PBC	Li metal	PPy/BC separator	$LiPF_6$	$LiFeO_4$	119.0 mAh g^{-1} (2 C)	[72]
CF-SO_3	Zn metal	CF-SO_3 separator	$ZnSO_4/MnSO_4$	MnO_2	~200.0 mAh g^{-1} (1 A g^{-1})	[46]
WBG	HC	WBG separator	$NaPF_6$	Na metal	>300.0 mAh g^{-1} (0.1 C)	[78]
CP	Li metal	CP-SSE film	LiTFSI	$LiFePO_4$	108.0 mAh g^{-1} (2 C)	[83]
CNC-PAN	Li metal	CNC-PAN film	LiTFSI	$LiFePO_4$	163.0 mAh g^{-1} (0.1 C)	[88]
ZCNF	Li metal	PL-ZCNF SSE	LiTFSI	$LiFePO_4$	152.7 mAh g^{-1} (0.5 C)	[85]
CMC	Zn metal	CMC hydrogel	$Zn(ClO_4)_2$	MnO_2	110.7 mAh g^{-1} (1 A g^{-1})	[86]

CMC: carboxymethyl cellulose. **LS**: sodium lignosulfonate. **HC**: hard carbon. **NM**: not mentioned. **CEF**: cellulose elementary fibrils. **OLO**: overlithiated layered oxide. **TOCNF**: TEMPO-oxidized CNF. **LiTFSI**: bis(trifluoromethane)sulfonimide lithium. **HEC**: hydroxyethyl cellulose. **NBC**: amino-grafted bacterial cellulose. **CLE**: cellulose levulinate ester. **PBC**: polypyrrole-coated bacterial cellulose. **PPy**: polypyrrole. **CF-SO_3**: sulfonated cellulose. **WBG**: woven fabric-based TEMPO-oxidized bacterial cellulose–glycine composite. **CP**: cellulose phthalate. **SSE**: solid-state electrolytes. **CNC-PAN**: cellulose nanocrystal grafted polyacrylonitrile. **BC**: bacterial cellulose. **ZCNF**: zwitterionic cellulose nanofiber. **PL**: PEO matrix and LiTFSI.

acteristics. Its inherent polar functionalization enables strong interfacial interactions with active materials, rendering cellulose an effective binder and dispersant for freestanding electrodes. Furthermore, cellulose-derived ultrathin membranes mitigate issues associated with fragile SEIs; their aligned nanochannels and tailored interfaces regulate ion deposition and suppress parasitic reactions. The mechanical resilience of cellulose, which originates from its covalent and hydrogen bonding, combined with hydroxyl-derived hydrophilicity, positions it as a dimensionally stable porous barrier that enhances ion transport. Cellulose also provides mechanical reinforcement while accommodating ion transport pathways in solid and quasi-solid electrolytes by virtue of its processability and three-dimensional network-forming ability. However, considerable challenges persist in harnessing the full potential of cellulose for high-performance energy storage applications. The following sections present a general outlook and outline potential future research directions (Fig. 7).

Advanced analytical methodologies. Despite its unique properties, the application of cellulose in energy storage systems is still in its infancy. The inherent variability in characteristics owing to source-dependent structural fluctuations and inconsistencies in production methodologies leads to unpredictable electrochemical performance of cellulose derivatives in energy storage systems. Consequently, adopting advanced and nondestructive analytical methodologies is required to advance cellulose research. Elucidating structural evolution patterns and dynamic interactions across varied operational environments would allow establishing precise structure–property–function relationships, which would promote material optimization strategies.

Artificial intelligence (AI)-driven methodologies. The integration of AI into energy storage innovation demonstrates considerable potential for accelerating technological advancement through enhanced data analysis and predictive modeling. Conventional experimental approaches relying on iterative physical testing often encounter efficiency constraints regarding temporal and economic resource allocation. AI-driven methodologies offer alternative pathways by enabling predictive material screening and computational optimization using machine learning algorithms. Although these techniques are particularly promising for addressing complex biopolymer applications where structural diversity complicates property prediction, current implementations remain constrained by insufficient training datasets and underdeveloped theoretical frameworks. Progress in this specialized domain requires systematic refinement of neural network architectures coupled with adaptive learning protocols to establish reliable correlations between molecular configurations and functional characteristics, which are essential for biomaterial engineering for energy storage applications.

Advanced battery systems. Conventional flexible batteries are typically constructed using a layered sandwich structure with an anode/cathode-separating membrane. This design often suffers from interfacial displacement and delamination under mechanical stress due to poor structural integration, severely increasing contact resistance and impeding ion transport. Integrated monolithic designs overcome these issues by creating unified systems with continuous interfacial connections. To realize such designs, cellulose is a prime candidate owing to its inherent

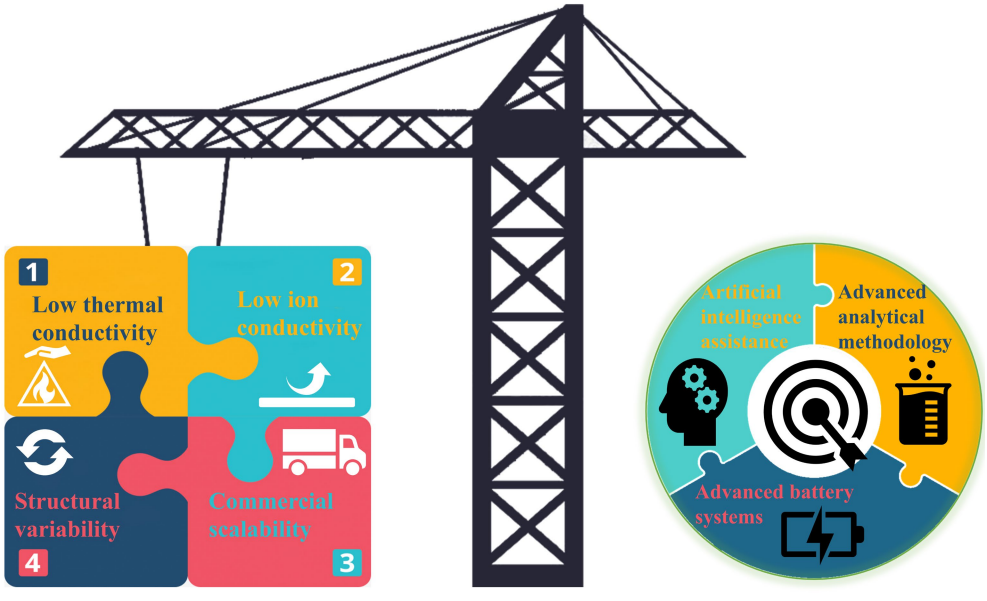

Figure 7 Challenges and strategies for the application of cellulose in energy storage devices.

hierarchical structure, superior film-forming ability, robust stability, and abundant reactive sites for functionalization. Furthermore, mechanically robust nanocellulose exhibits enhanced functional properties, boosting the electrochemical performance. These characteristics render cellulose-based materials highly promising for developing stable, high-performance integrated battery systems.

In summary, cellulose materials hold remarkable promise for enhancing the performance and sustainability of energy storage devices. Coupling its unique features with advanced emerging technologies can be expected to provide a powerful platform for further development of electrochemical energy systems.

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Author contribution statement

Longjun Chang is responsible for writing the original drafts. Zhaoxin Li and Zhiyuan Yan contribute greatly to the review and revision of the article. Qing Li takes the oversight and leadership responsibility for the whole process. All the authors have approved the final manuscript.

Data availability

Not applicable.

Declaration of competing interest

All the contributing authors report no conflicts of interest in this work.

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Use of AI statement

None.

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