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Toward More Recyclable and Sustainable Lithium-Ion Batteries

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ABSTRACT: The rapid rise of electric vehicles has driven significant advancements in lithium-ion battery (LIB) technology. However, the pursuit of higher power, longer lifespan, and greater capacity has often overshadowed critical factors such as recyclability, material sustainability, and environmental impact. To ensure the long-term viability of LIBs, future developments must strike a balance between performance and sustainability. This review advocates for the design of next-generation battery components using abundant, environmentally friendly, and sustainable materials that minimize reliance on intensive mining and refining processes. Key innovations include metal-free electrodes, nonmetallic current collectors, solid-state electrolytes, biodegradable components, anode-free configurations, and water-soluble binders for electrode fabrication. We provide a comprehensive assessment of recent progress in these areas, discuss challenges for practical implementation, and outline future directions for advancing these technologies. By offering a forward-looking perspective on innovative battery materials and designs, this review provides valuable insights into the sustainable evolution of LIBs.

KEYWORDS: *electric vehicles, spent lithium-ion batteries, environmental impacts, sustainable battery design, ease of recyclability*

INTRODUCTION

The widespread adoption of electric vehicles (EVs) has resulted in significant demand growth of lithium-ion batteries (LIBs).[1] As of 2022, nearly 30 million EVs are in operation globally, with projections indicating this figure could reach 250 million by 2030.[2] In addition, 77% of the electricity power storage systems employed for grid stabilization rely on LIBs technology.[3] This growing reliance on LIBs technology raises concerns about the sustainable supply of critical materials, such as lithium (Li), cobalt (Co), nickel (Ni), and manganese (Mn). By 2022, approximately 60% of Li, 30% of Co, and 10% of Ni mineral resources have already been consumed.[4] The current linear development model of LIBs, from mining and manufacturing to application, inevitably causes resource depletion and supply chain risks (Figure 1a).[5] Thus, a shift from a linear to a circular development model is strongly recommended (Figure 1b). In this circular model, the LIBs lifecycle is closed through efficient recycling of spent batteries, which not only alleviates resource scarcity issue, but also mitigates the environmental impact of LIB disposal, promoting the sustainable development of LIBs.[6−8]

Regrettably, to date, less than 6% of spent LIBs have been recycled due to recycling technology limitations, regulatory gaps, and poor traceability, particularly given the insufficient emphasis on recyclability during the manufacturing of LIBs.[9,10] Traditional LIBs, which comprise cathodes containing heavy metals, organic separators, and toxic electrolytes, can cause severe environmental damage if not properly disposed of when they reach the end of their lifecycle.[11] Currently, the common methods for processing spent LIBs listed in Table 1 are faced with significant challenges, including high energy consumption, substantial greenhouse gas (GHG) emissions, and most critically, low recycling efficiency.[12,13] Disassembling batteries into their individual components presents a more effective way to enhance recycling efficiency; however, the complex structure and materials of LIBs complicate the precise separation of components.[14] For example, the presence of metal collector fragments, robust poly(vinylidene fluoride) (PVDF) binder and highly stable graphite in black mass makes it extremely difficult to obtain pure cathode powder for further efficient recycling or regeneration.[15] Black mass refers to the powdered mixture of battery components resulting from mechanical crushing and shredding. Therefore, it is essential to prioritize battery materials and designs that facilitate easy, smart, and sustainable recycling of LIBs.

In this review, we start with a rigorous analysis of the environmental impacts of the components in traditional LIBs, and reveal that the manufacturing of cathodes contributes the

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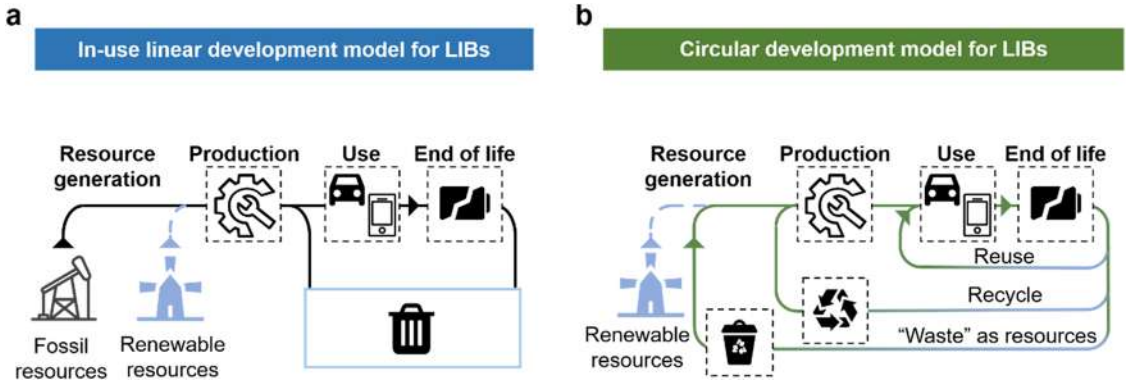

Figure 1. (a) Linear development model for LIBs; (b) circular development model for LIBs.

most to total energy consumption and GHG emissions. Following cathodes, the environmental footprint of anodes, current collectors, electrolytes, and separators is also significant, primarily due to the extensive mineral mining and refining processes required for their manufacturing. To lessen the reliance on mineral resources in LIBs development, we advocate for the exploration of abundant, safe, and sustainable alternatives to conventional components. These include metal-free electrodes, nonmetallic current collectors, solid-state electrolytes, biodegradable materials, anode-free designs, and water-soluble binders for electrode fabrication. This review encapsulates recent progress in these pivotal areas, acknowledges existing challenges, and outlines future research directions aiming for practical applications of these innovations. A visual representation of our perspective is shown in Figure 2a, highlighting the essential steps toward a sustainable battery future. Some key metrics of these sustainable batteries are compared with conventional LIBs, which is illustrated in Figure 2b. Although some of these batteries are inferior to traditional batteries in terms of capacity, cycling life or energy density, they show clear advantages in terms of material sustainability. Future research can focus on improving their performance.

The Environmental Impacts of Traditional LIBs. Taking the manufacturing of $LiNi_{0.33}Co_{0.33}Mn_{0.33}O_2$ (NCM111) battery as an example, Dai et al. analyzed the energy consumption, water use and GHGs emissions associated with each component in the production of a 1 kWh battery (Figure 3).[16] Their findings show that the production of the cathode is the most energy-intensive component, consuming 410 MJ and 238 L of water, while emitting 28.5 kg of GHGs. This accounts for 36.4% of the total energy consumption, 31.7% of water use, and 39.1% of GHG emissions, respectively.[17] In contrast, while the production of synthetic graphite does not require excessive energy, it releases significant amounts of nitrogen oxides (NO_x), sulfur oxides (SO_x), and particulate matter (PM10) due to the combustion of impurities found in coal tar pitch and petroleum coke.[18] Additionally, the manufacturing of aluminum (Al) and copper (Cu) current collectors substantially contributes to the energy consumption and GHG emissions, particularly SO_x emissions, primarily due to the processing and refining of copper sulfide ores.[19] Furthermore, the production of electrolyte, organic salt ($LiPF_6$), separator, and the whole cell also consume considerable energy and generate large amount of GHGs. Similarly, in the production of $LiFePO_4$ (LFP) and $LiMn_2O_4$ (LMO) batteries, cathode production remains notably energy-intensive compared to other components, with Freire et al. reporting energy consumption of 529 MJ/kWh for LFP and 304 MJ/kWh for LMO cathode materials.[20] Therefore, it has become imperative to reimagine battery materials and structure to ensure more sustainable practices, with proposed solutions to be discussed in detail in the following sections.

Metal-Free Electrode Materials. Given the substantial environmental impact of traditional LIB cathodes, it is essential to explore alternatives that do not rely on critical metals. One promising approach is the use of metal-free materials, such as organic electrode materials. These materials are primarily composed of abundant elements such as carbon (C), hydrogen (H), oxygen (O), nitrogen (N), and sulfur (S), which can be sourced from biomass through various reaction pathways. Furthermore, the characteristics of these electrode materials, such as chemical potential, stability, and electrochemical performance, can be tailored by incorporating functional elements during chemical transformation to meet the specific needs of various battery applications. Crucially, at the end of the battery's lifecycle, these organic electrode materials can naturally degrade into environmentally benign substances, leaving no harmful residuals.

Extensive research on organic electrode alternatives has primarily focused on materials such as conductive polymers,[21] organosulfur compounds,[22] organic radicals,[23] conjugated carbonyl compounds,[24] and imine/azo compounds.[25] Figure 4a compares the characteristics of these organic electrode materials. Conductive polymers demonstrate reasonably high electronic conductivity but suffer from low Coulombic efficiency and a narrow voltage window.[26] Organosulfur compounds typically offer high capacity; however, they exhibit significant electrochemical polarization and slow electrochemical kinetics.[27] Organic radical species, such as 2,2,6,6-tetramethylpiperidin-1-oxyl (TEMPO), 2,2,5,5-tetramethylpyrroline-*N*-oxyl, and nitronyl nitroxide units,[26,28] provide relatively high discharge voltages. However, their limited electron transfer stoichiometry restricts discharge capacity to around 100 mAh g^{-1} or lower. Carbonyl compounds (quinones, carboxylic acids, anhydrides, imides, and ketones) have garnered extensive research interest owing to their augmented capacity and fast electrochemical kinetics.[29] However, small carbonyl compounds often face solubility challenges in nonprotonic electrolytes. While using aqueous electrolytes can mitigate the dissolution of these materials, the discharge products still tend to dissolve in water, leading to rapid capacity degradation.[30] In contrast, large conjugated systems with multiple carbonyl functional groups show promising features, including high stability, enhanced specific capacity, fast reaction kinetics, and structural diversity.[31,32] These properties make them strong candidates

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Table 1. Comparison of Different Recycling Methods

method	process steps	advantages	disadvantages
pyrometallurgy	high-temperature incineration (800−1500 °C) decomposes organics; metals enriched as alloys/oxides	high adaptability, handles complex batteries, high metal purity (>95% for Co, Ni)	high energy consumption, emits toxic gases (e.g., HF, dioxins), requires gas treatment systems
hydrometallurgy	acid/alkali leaching (e.g., H_2SO_4, HCl) dissolves metals, followed by precipitation/solvent extraction/electrowinning	high metal recovery rate (>90%), ultrahigh purity (>99%), scalable for industrial use	generates acidic/alkaline wastewater, complex chemical processing, high reagent costs
direct regeneration	Li replenishment, thermal treatment, and electrochemical reactivation to restore cathode materials (e.g., $LiCoO_2$, $LiFePO_4$)	low cost (30−50% cheaper than hydrometallurgy), high material recovery (>95%), low carbon footprint	only viable for lightly degraded batteries, requires precise sorting, immature industrial technology

for next-generation high-performance, eco-friendly, and sustainable LIB cathodes.

It is worth noting that the low density of organic electrode materials (1 g cm^{-3} or less) compared to traditional cathodes (3.6−5.1 g cm^{-3}, Figure 4b), results in a lower volumetric energy density. In addition, their low electronic conductivity remains a major limitation. Figure 4c compares the conductivities of typical inorganic electrode materials and selected organic electrode materials. Materials like LCO, LMO, and LFP, show conductivities of 10^{-4}, 10^{-6}, and 10^{-9} S cm^{-1}, respectively, and graphite has a much higher conductivity of around 10^{4} S cm^{-1}. However, most organic electrode materials, except for conductive polymers, are not sufficiently conductive. For instance, organosulfur compound has a conductivity of 5.9×10^{-13} S cm^{-1},[33] and organic radicals exhibit approximately 5×10^{-11} S cm^{-1}.[34] Sulfur shows an extremely low electronic conductivity of around 10^{-30} S cm^{-1}. Therefore, significant efforts are needed to improve the electronic conductivity of organic electrode materials to make them viable alternatives to existing electrodes.

The working principle of organic electrode materials can be classified into three distinct types, as illustrated in Figure 4d.[26] N-Type organic molecules (such as quinones) undergo reduction during the discharge process, forming anions that then complex with Li^+. Since most N-type organic materials lack Li^+, they need to be paired with either Li-containing cathodes (configuration I) or Li-containing anodes (configuration II). P-type organic materials (such as sulfur ethers) can interact with anions in electrolytes (e.g., PF_6^-, ClO_4^-) upon oxidation and can be coupled with conventional low-voltage anodes like graphite (configuration III). Chen et al. conducted a comparative analysis of four representative organic electrode materials in full cells paired with either a $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ (NCM622) cathode or Li metal to assess their practical application potential.[35] The molecular structures of the four organic electrode materials are illustrated in Figure 4e. They set the total energy and power of each battery system at 100 kWh and 150 kW, respectively. The results revealed that substituting graphite (G) with compounds (A) or (B) anode significantly reduced the gravimetric and volumetric densities, while also increasing material costs compared to the NMC622-G battery (Figure 4e). In contrast, the (C)-Li battery achieved an energy density of 268 Wh kg^{-1}, approaching that of the NMC622-Li battery, while its cost was $100 per kWh, much lower than other organic battery options. Consequently, batteries based on p-type organic electrode materials with high redox potential (particularly carbonyl compounds) demonstrate substantial potential for practical application.

Despite the great promise of organic electrode materials for sustainable LIBs, their large-scale adoption faces a few challenges. First, the inherent poor electronic conductivity of organic electrode materials necessitates the use of a substantial amount of conductive carbon additives, which not only raises costs but also lowers battery's specific energy density. Improving the conductivity of organic materials must therefore be a central focus of future research,A recent study demonstrated that the conductivity of Carbon Nanotubes (CNTs) could be enhanced by incorporating polyaniline (PANI) via a solution-based method. This approach suggests that a similar strategy might be feasible for improving the conductivity of organic electrode materials.[36−38] Additionally, the performance evaluations of organic electrode materials are often conducted under laboratory conditions that do not reflect real-world applications.

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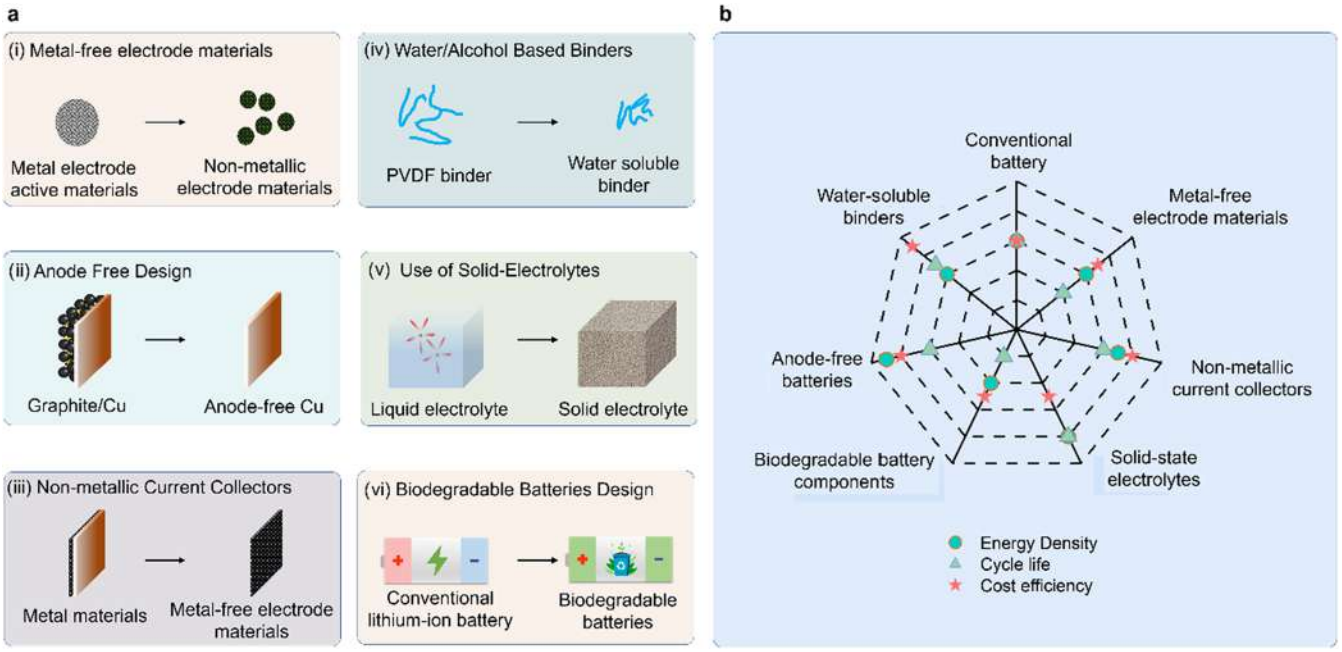

Figure 2. (a) Trajectory demonstrating solutions for next-generation, recyclable, and sustainable LIBs; (b) comparison between traditional batteries and next-generation sustainable batteries.

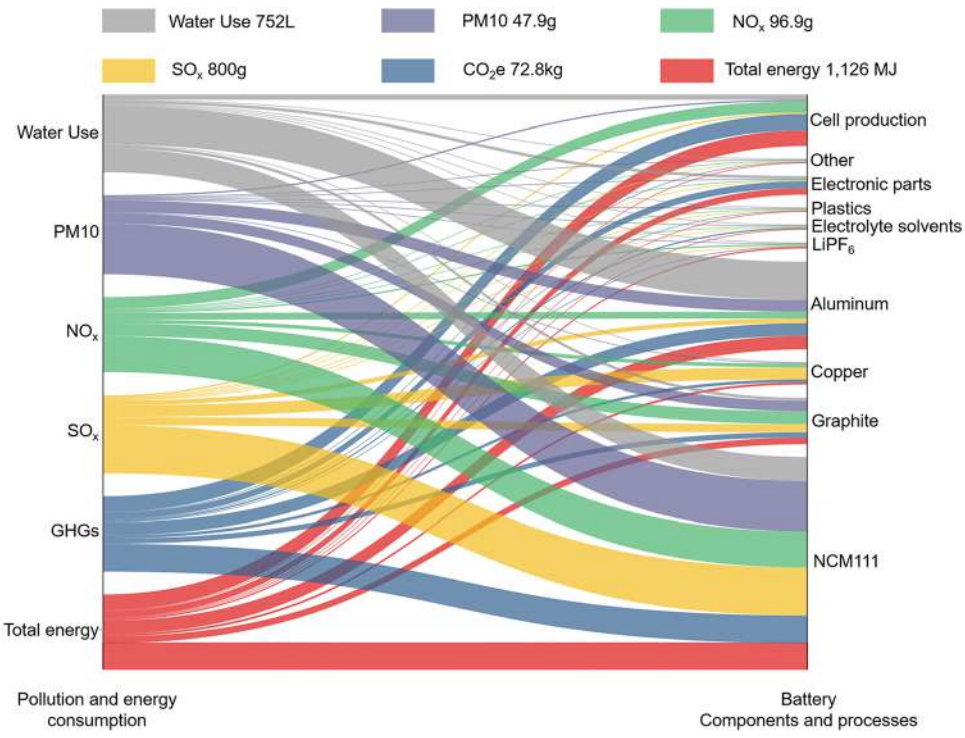

Figure 3. Cradle-to-gate impact breakdowns and bill of materials (BOM) for manufacturing 1 kWh NCM111 battery.[16]

Parameters such as mass loading, electrolyte volume, and cathode-to-anode ratios often differ from industry standards, making it crucial to test these materials under conditions that closely mirror practical usage. Finally, most organic materials are not yet produced at the scale of commercial inorganic materials, limiting their immediate availability and competitiveness. To transition from academic research to practical use, these challenges must be addressed by improving conductivity, aligning testing protocols with real-world conditions, and developing scalable, cost-effective production methods for organic electrode materials.

Nonmetallic Current Collectors. Commercial LIBs typically use Al and Cu as current collectors for cathode and anode electrode, respectivly.[39] However, these metallic collectors contribute significantly to the overall weight of LIBs, reducing their gravimetric energy density. Additionally, the complete separation of Al and Cu from spent battery black mass poses significant challenges, reducing both recycling efficiency and the purity of recovered products.[40] In contrast,

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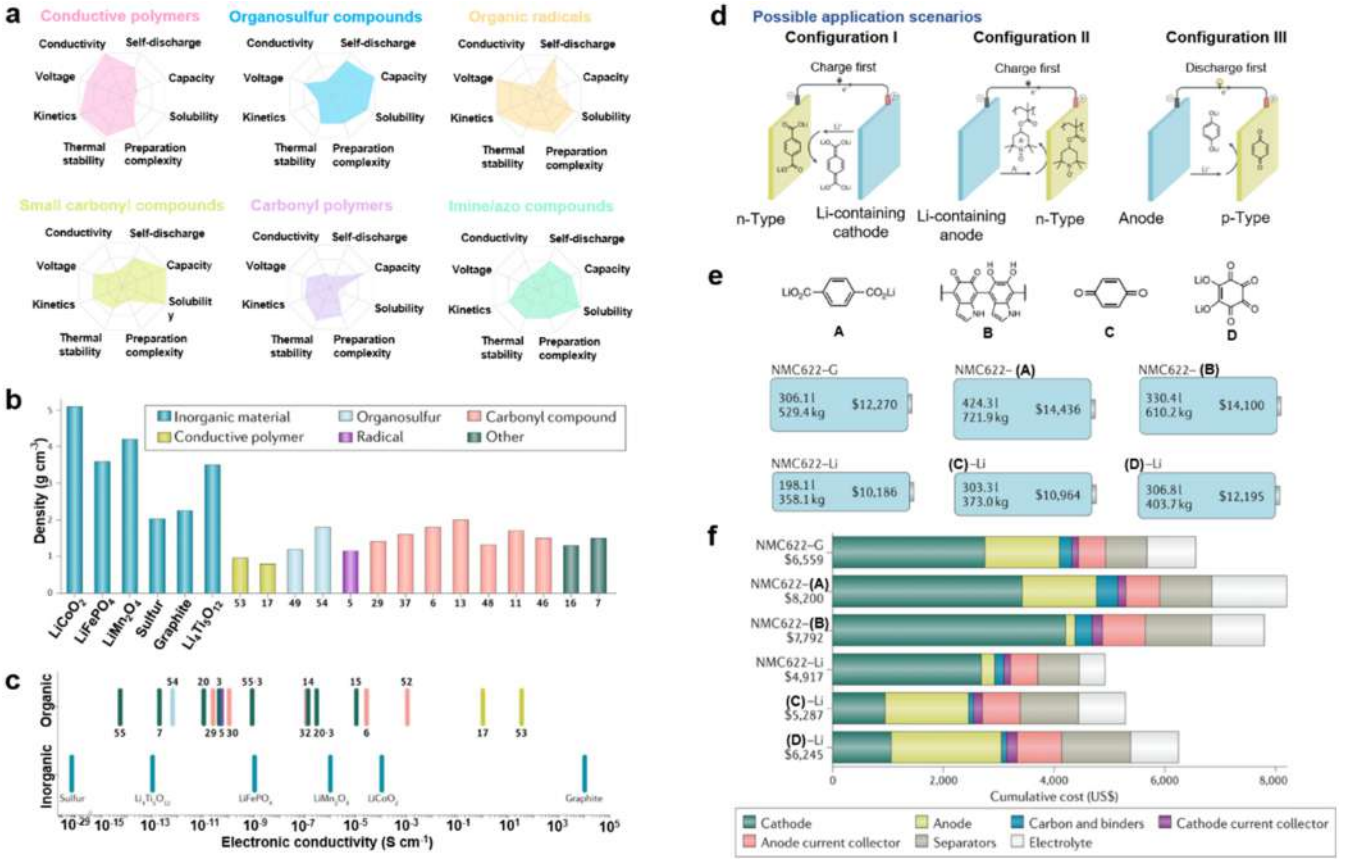

Figure 4. (a) Overview of the fundamental properties of different types of organic electrode materials;[27] Adapted with permission from ref27. Copyright 2018 Elsevier. (b) Gravimetric density and (c) electronic conductivity of different organic and inorganic electrode materials;[35] The numbers in this figure correspond to the organic compounds reported in ref 35. (d) Three configurations for LIBs based on organic electrode materials;[35] (e) molecular structures of four representative organic electrode materials and total volume and mass of the assembled battery systems, as well as the associated manufacturing costs;[35] (f) cost breakdown of materials used in the assembled battery systems.[35] Adapted with permission from ref35. Copyright 2020 Springer Nature.

carbonaceous materials with high electrochemical stability offer a promising alternative for current collectors in LIBs. These materials are lightweight (density around 0.44 g cm^{-3}) compared to Al (2.7 g cm^{-3}) and Cu (8.96 g cm^{-3}). Replacing only Cu foil could reduce the total weight of an LIB by 12%, resulting in a 14% increase in energy density.[41] Furthermore, the mechanical flexibility of carbon-based current collectors could broaden the application of LIBs in portable electronic devices.[42]

However, traditional wet-coating processes may not be applicable for carbon-based current collectors due to their porous and soft nature.[43] Especially during the drying phase, solvent evaporation can lead to uneven stress, causing the edges of the current collector to fold inward.[43] To address this challenge, Kim and his team developed a method for preparing electrodes using a carbon fabric current collector (Figure 5a). In their approach, a slurry of LFP was first applied to a glass substrate at a specified thickness, followed by the placement of carbon fabric on top. The entire assembly was then placed into a vacuum bag autoclave with porous silicon rubber. During the autoclaving process, the evaporated solvent permeated through the fabric's pores, allowing the carbon fabric to maintain its original shape and structural integrity.[44]

In addition to advancements in coating techniques, many researchers have developed self-supporting electrode structures through vacuum filtration methods.[41] Guo et al. developed a process where they first dispersed ultralong, monodispersed CNTs in NMP solvent with the help of poly(vinylpyrrolidone) (PVP) and sodium cholate (SC) (Figure 5b). Following this, LFP cathode particles were distributed in the CNT dispersion. After vacuum filtration, a self-supporting LFP electrode was obtained (Figure 5c). This CNT-based robust network imparted excellent mechanical strength and conductivity. The free-standing LFP electrode achieved a high mass loading of 39.1 mg cm^{-2}, and could withstand a stress of up to 7.2 MPa and 5% strain.[45] Additionally, the electrode delivered a capacity of 139.1 mAh g^{-1} at a rate of 2C rate, outperforming traditional thick electrode with Al current collectors.[46] This work marks a significant advancement in the fabrication of binder-free and current collector-free electrodes.

In addition to the progress in cathodes, advances in self-supporting anode electrodes without Cu current collectors have also been made. Lu et al. developed a $Li_4Ti_5O_{124}Ti_{52}$ free-standing anode on carbon fiber current collector (LTO/CF) using a sequential vacuum filtration method (Figure 5d). This anode maintained its morphology, mechanical and electrochemical properties even after 4000 repeated bending cycles.[47] Due to the eliminating of metal current collector, the mass fraction of LTO in the electrode was increased from 17% to 91%, significantly enhancing the battery's gravimetric energy density. Furthermore, they used the same approach to fabricate LFP/CF electrode and assembled a full cell with the LTO/CF anode. Due to the use of a lightweight CF current collector, the final full cell

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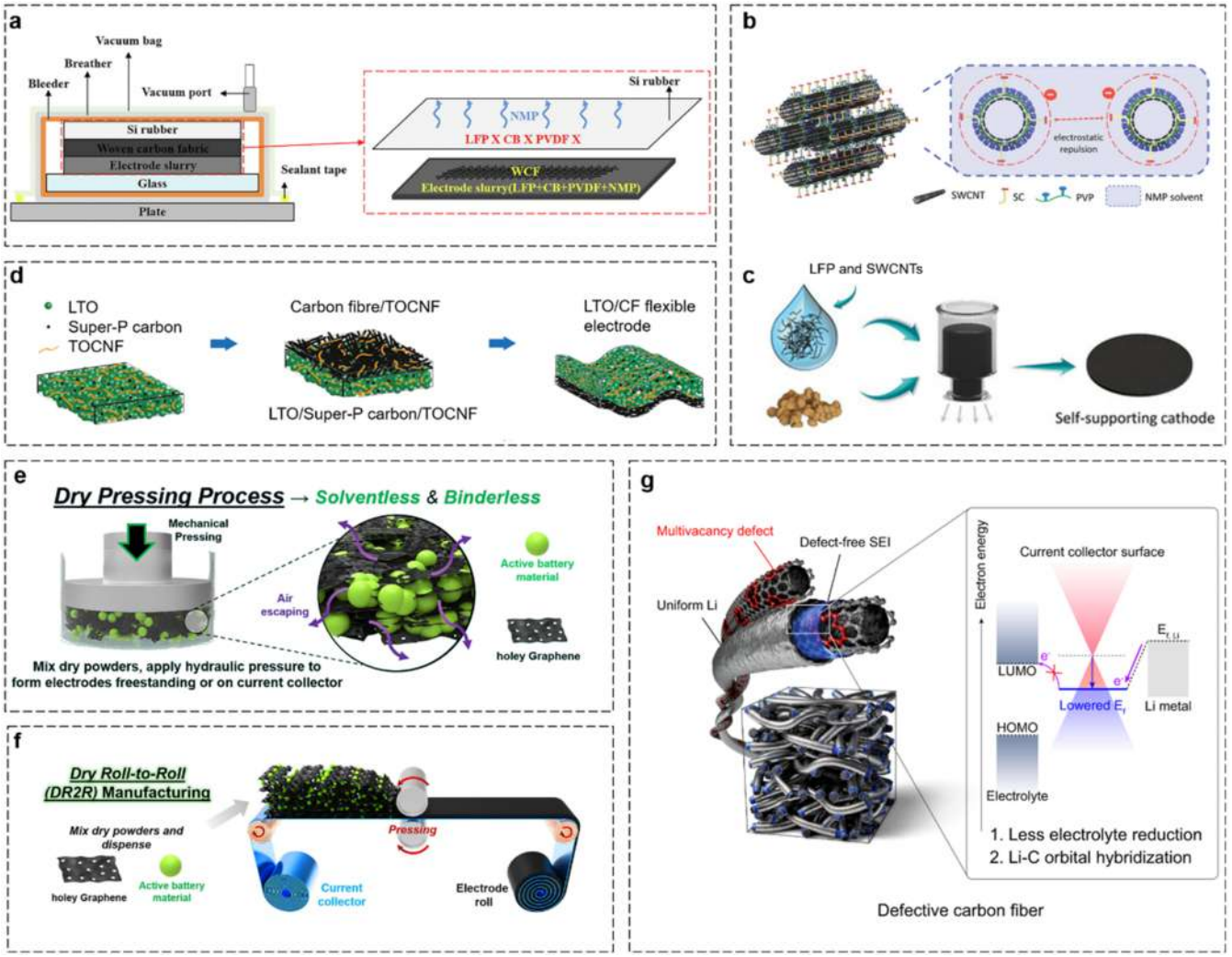

Figure 5. (a) Schematic illustration of the vacuum bagging electrode preparation method on carbon fabric current collectors;[44] Adapted with permission from ref 44. Copyright 2021 Elsevier. (b, c) The dispersion mechanism of monodispersed CNTs and the preparation flowchart for the self-supporting LFP cathode;[45] Reproduced from ref 45. Available under a CC BY license. Copyright YanQiang Wang. (d) Schematic illustration of the fabrication procedure for the LTO/CF electrode;[47] Adapted with permission from ref 47. Copyright 2021 Elsevier. (e) Schematic illustration of the dry pressing process for preparing binder-free and solvent-free self-supporting electrodes; (f) schematic illustration of pressing the self-supporting onto metallic current collector.[48] Adapted with permission from ref48. Copyright 2019 American Chemical Society; (g) schematic of the electron-deficient carbon paper current collector and its underlying mechanism for improving anode-free battery performance.[53] Reproduced from ref 53. Available under a CC BY license. Copyright Hee-Tak Kim.

showed a high energy density of 90 Wh/kg (Compared to the conventional LFP battery with an energy density of 109 Wh/kg).

Additionally, Hu et al. developed a dry processing method to prepare substrate-free electrodes.[48] By integrating holey graphene (hG) with active materials using hydraulic technology, they eliminated the need for binders and solvents during the electrode preparation (Figure 5e).[48] After cold pressing, the active material particles were securely embedded within the 3D conductive network of hG. The resulting LFP electrode exhibited an impressive initial capacity of 160 mAh g^{-1}. However, the weak mechanical properties of these self-supporting electrodes posed challenges for compatibility with existing battery production processes. As a result, the authors suggested that large-scale production might require pressing the free-standing electrodes onto current collector using a roll-to-roll method to ensure acceptable performance. (Figure 5f).[48] At the Tesla Battery Day event in 2020, the company announced the development of 4680 cylindrical cells fabricated using freestanding dry-processed anodes, marking a significant advancement in electrode manufacturing technology.[49]

Notably, carbon-based current collectors also demonstrate outstanding performance in anode-free LIBs (AFLIBs), as their 3D structure can effectively suppress the growth of Li dendrites and enhances battery performance.[50,51] Qu et al. prepared a graphene array on a graphene film, which was directly used as the current collector of AFLIBs. The high conductivity and pore-rich structure of the electrode enabled rapid Li^+ diffusion and efficient charge transport, resulting in a high capacity of 770 mAh g^{-1}, which outperforms commercial graphite anodes (with a capacity of approximately 372 mAh g^{-1}).[52] Wan et al. developed a lightweight, flexible, and free-standing 3D hollow carbon fiber (3D-HCF) current collector, which showed an average Coulombic efficiency of up to ~99.5% over 350 cycles and a cycle life of more than 1200 h due to the reduced local current density and suppressed Li dendrite growth.[50]

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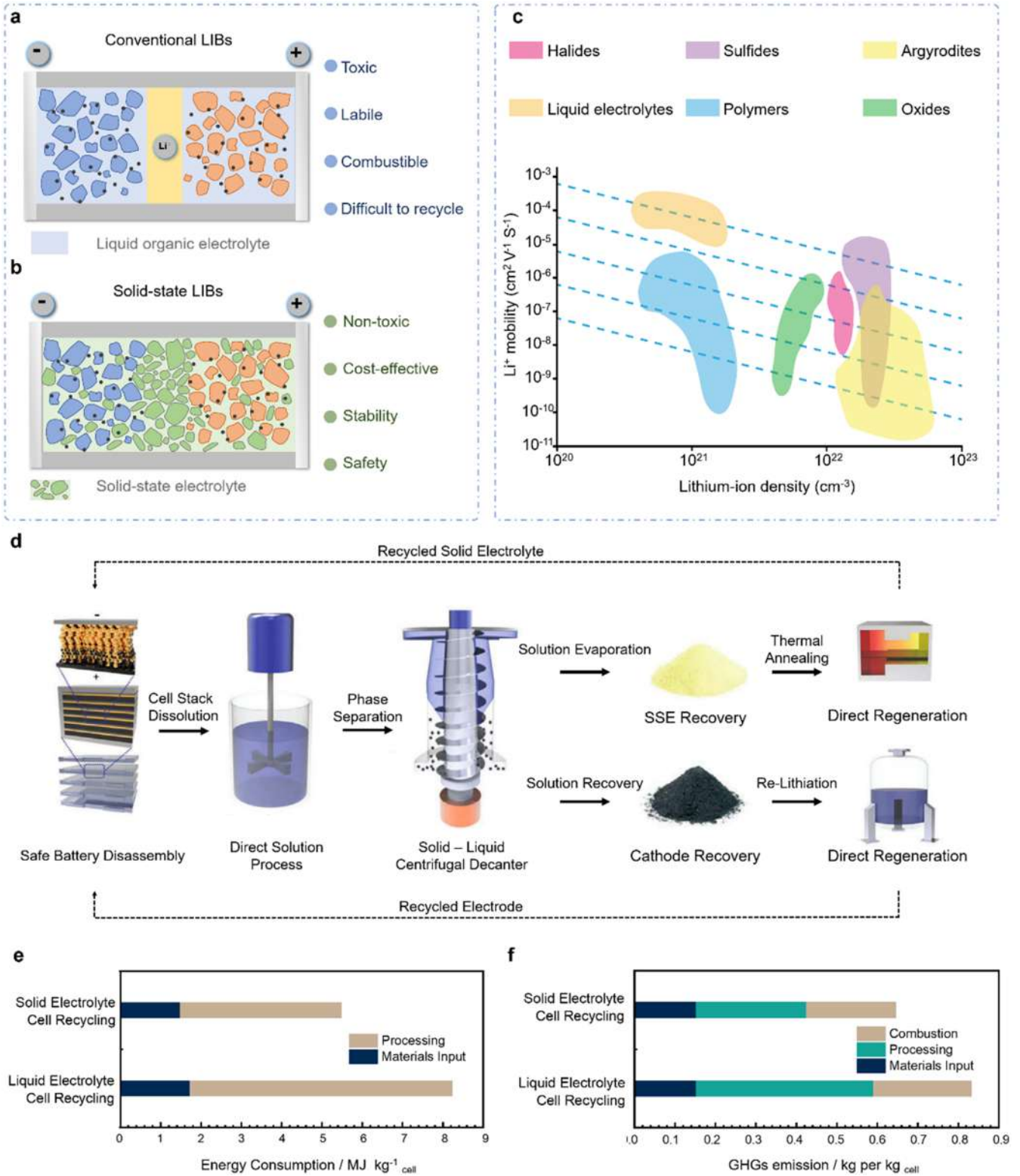

Figure 6. (a, b) Structure of conventional LIBs and ASSLIBs; (c) the state-of-the-art SSEs; (d) schematic of the spent ASSLIBs recycling procedure;[72] (e, f) energy consumption and GHGs emission of recycling batteries based on LEs and SSEs.[72] Reproduced from ref 72. Available under a CC BY license. Copyright Zheng Chen.

Kim et al. directly employed carbon paper as the current collector in AFLIBs but encountered difficulties in forming a uniform solid electrolyte interface (SEI). To address this, they introduced multivacancy defects, creating electron-deficiencies on carbon paper substrate (Figure 5g). These defects strongly could bond with Li^+ via orbital hybridization, promoting uniform Li deposition. Additionally, the reduced Fermi level of the modified carbon paper minimized electrolyte reduction, resulting in a thin, uniform SEI layer. Remarkably, the NCM811/carbon paper AFLIB assembled by Kim et al. retained

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90% of its initial capacity after 50 cycles, demonstrating good cycling stability.[53]

However, the practical application of carbon-based current collectors in LIBs remains challenging. Traditional electrode processing methods are not well-suited for these collectors. While researchers have developed advanced methods such as vacuum bagging, filtration, and hydraulic pressing, these methods are primarily limited to laboratory settings and are difficult to scale for commercial production. Scaling these processes without compromising performance or increasing costs is a significant hurdle. Furthermore, carbon-based current collectors generally have lower mechanical strength than their metallic counterparts, making them less durable under mechanical stresses during battery manufacturing and operation. This issue is particularly problematic during long-term cycling, where poor mechanical properties can lead to rapid capacity degradation of LIBs. Further research could focus on enhancing the mechanical properties of carbon-based current collectors through composite design (e.g., incorporating transition metal oxides/sulfides/nitrides, MXenes, or metal−organic frameworks (MOFs)[54−58]), improved processing techniques, and hybrid systems that combine the advantages of both carbon and metal-based current collectors.

Solid-State Electrolytes. Traditional liquid electrolytes (LEs) used in LIBs typically consist of organic solvents such as ethylene carbonate (EC), dimethyl carbonate (DMC), diethyl carbonate (DEC), combined with organic Li salts such as $LiPF_6$. These components are flammable, explosive, and toxic, posing significant challenges for both the manufacturing and recycling of LIBs.[59,60] For example, the hazardous nature of LEs requires that spent LIBs be mechanical crushed in an inert atmosphere and fitted with waste gas purification facilities to comply with emission standards, which drives up recycling costs.[61] Although techniques like supercritical CO_2 extraction for electrolyte recycling have been proposed, high costs, limited profitability, and low purity of the recycled electrolytes hinder widespread adoption. In contrast, shifting from LEs to solid-state electrolytes (SSEs) can significantly enhance battery safety and performance while facilitating safer recycling of spent LIBs (Figure 6a,b).

The state-of-the-art SSEs include sulfides, oxides, halides, and polymers (Figure 6c).[62] Sulfide SSEs, such as $Li_{10}GeP_2S_{12}$(LGPS) and $Li_6(P,Sb)S_5X$ (where X represents halogens), exhibit superionic conductivities exceeding 1 mS cm^{-1} and good mechanical ductility. However, they face challenges with cathode-electrolyte interfacial stability and moisture instability.[63] Oxide SSEs generally exhibit high stability and environmental compatibility, but their low conductivity and insufficient solid−solid contact limit their applications.[64] Halide SSEs are recently considered as appealing candidates due to their favorable balance of electrochemical stability (>4 V versus Li^0), ionic conductivity (0.5−12 mS cm^{-1}) and mechanical deformability, which allows for stable operation with conventional oxide-type cathodes.[65,66] However, halide SSEs are prone to hydrolysis in humid conditions, which reduces their ionic conductivity and limits their applications.[67] Polymer SSEs offer a promising solution to the challenge of achieving good solid/solid contact between electrodes and solid electrolytes.[68] They typically exhibit high mechanical flexibility and favorable interfacial wettability, creating a compact solid/solid interface that can accommodate volume changes during charging and discharging cycles. However, the main limitation with solid polymer electrolytes lies in their narrow electrochemical window. Poly(ethylene oxide) (PEO) can be rapidly oxidized at voltages above 4.2 V (vs Li^+/Li).[69]

Notably, when developing SSEs, it is crucial to prioritize not only their performance but also their recyclability. This not only reduces dependence on rare minerals like Ge and Ta during manufacturing,[70] but also lowers the overall production costs of all-solid-state LIBs (ASSLIBs). Zhou et al. assessed the feasibility of various recycling methods for spent ASSLB.[71] In pyro-and hydrometallurgical recycling processes, SSEs cannot be recycled, often being converted into alloys or metal salts, which can negatively impact the efficiency of extracting valuable metals. In contrast, direct recycling holds great promise for the recovery of SSEs while reducing the environmental impact of the recycling process.

Recently, Chen et al. successfully demonstrated direct regeneration of spent ASSLIBs.[72] They utilized the differing solubility properties of the sulfide-based electrolyte (Li_6PS_5Cl) and $LiCoO_2$ (LCO) in ethanol to facilitate the preseparation of the SSEs and cathodes. Following solvent evaporation and thermal annealing, the SSE was reusable. After hydrothermal treatment and short sintering, the spent LCO was regenerated. Notably, batteries assembled with the recycled electrolyte and cathode performed comparably to those made with pristine materials. Furthermore, the life cycle assessment (LCA) analyses indicated that recycling solid electrolyte cells produced lower GHGs and energy consumption compared to recycling liquid electrolyte cells (Figure 6e,f).

In addition to sulfide-based SSEs, several regeneration approaches have been explored for halide SSEs. One method involves dissolving the SSEs in a suitable solvent, followed by the addition of a Li salt to facilitate relithiation. The solvent is then evaporated to precipitate the regenerated SSEs.[73] Another method incudes high-energy ball milling or thermal treatment of cycled SSEs with additional Li salts to resynthesize SSEs.[74,75] Regeneration of polymer-based SSEs, such as poly(ethylene oxide) (PEO), has also been reported. In these cases, a simple water treatment can dissolve PEO into monomers within 30 min.[76] After water evaporation, Li salts and monomers can be recovered.

However, regeneration of oxide-based SSEs presents a challenge due to their similarities with oxide cathodes, making their separation difficult. Huang et al. developed an environmentally friendly deformation-driven resintering (DDR) route to recycle $Li_{6.5}La_3Zr_{1.5}Ta_{0.5}O_{12}$ electrolyte. The DDR technique uses mechanical forces to fracture the spent SSEs into smaller particles, enhancing Li^+ absorption during regeneration. They demonstrated that the ASSLIBs assembled with recycled SSEs and LFP cathode, exhibited a discharge capacity of 126.7 mAh g^{-1} and maintained 89.7% after 400 cycles at 0.5C. However, the study did not mention how the SSE and cathode were separated.[77]

In summary, among the various types of SSEs, polymer-based SSEs hold great promise for the sustainability of ASSLIBS. Their inherent differences from oxide cathodes allow for easier separation from cathode materials, facilitating following regeneration of electrolytes and cathodes. Additionally, polymer-based SSEs offer benefits such as low cost, ease of processing, and scalability, which further enhances their potential for widespread adoption in future battery technologies. However, several key challenges must be addressed for practical applications. The ionic conductivity of polymer-based SSEs is still lower compared to other SSEs like sulfides and oxides, limiting their performance in high-power applications. Fur-

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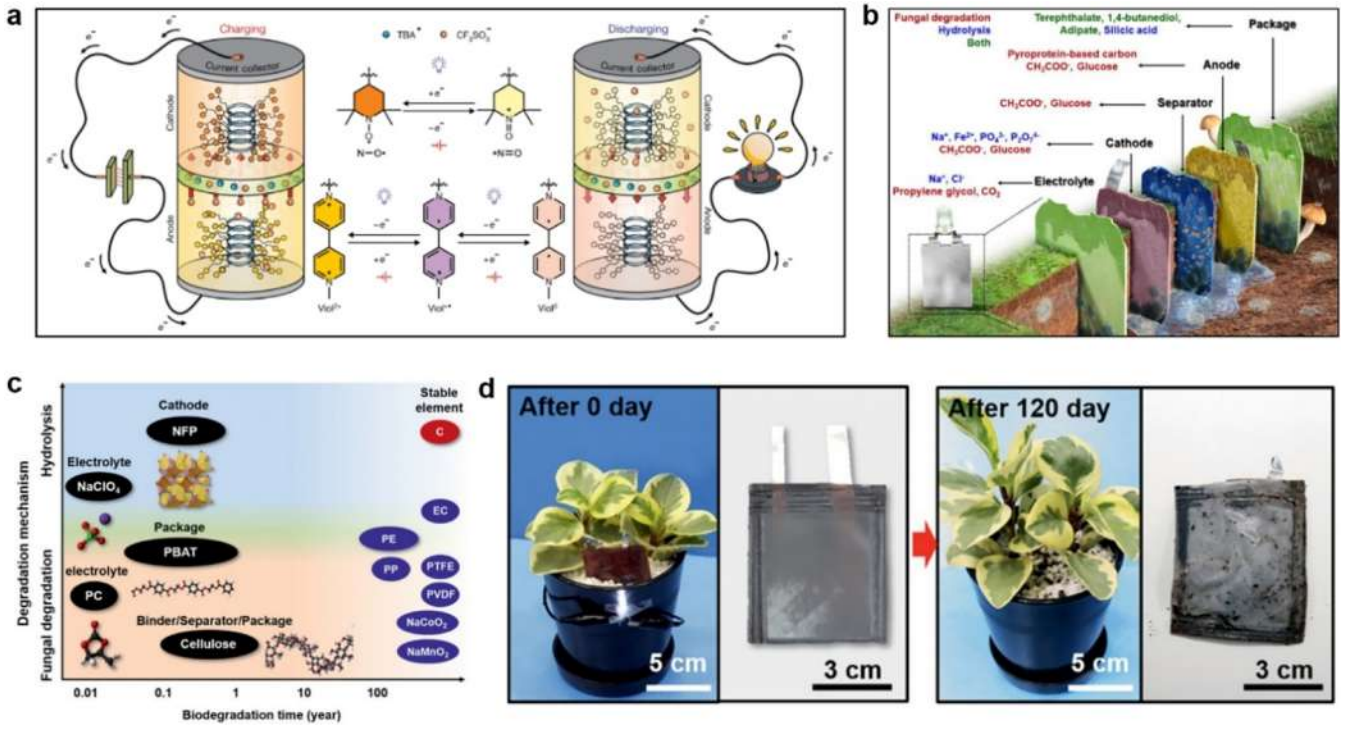

Figure 7. (a) Schematics illustration of the polypeptide-based organic radical battery and the involved electrochemical reactions that occur during charging and discharging process;[81] Adapted with permission from ref81. Copyright 2021 Springer Nature. (b) Components of the fully biodegradable SIBs and their degradation products; (c) biodegradation mechanism and timeline for different battery components;[83] (d) images of the plant before and after burying the used NIBs for 120 days.[83] Adapted with permission from ref83. Copyright 2021 John Wiley and Sons.

thermore, their temperature stability needs significant improvement to ensure safe operation under a wide range of conditions, particularly for EVs and grid-scale storage. Compatibility with electrodes, especially Li metal anodes, also requires further research to address issues such as dendrite formation and poor interfacial contact. Facing the above challenges, some companies appear to have found potential solutions. For instance, BYD has announced plans to adopt ASSLIBs in electric vehicles by 2027.[78] Tesla has also shown relevant developments in this area, suggesting that the widespread adoption of solid-state batteries may be realized in the near future.

Biodegradable Battery Components. Conventional LIBs components contain hazardous elements that can take a century to decompose if landfilled, causing serious pollution to soil and water bodies. To address this issue, recent studies have explored the use of biodegradable alternatives for electrode materials, binders, and separators, etc., which could potentially reduce the environmental impact of LIBs.

For instance, Guo et al. developed a biodegradable LIBs using emodin (6-methyl-1,3,8-trihydroxyanthraquinone) as the cathode and lithiated humic acid as the anode.[79] This battery exhibited a capacity of 157 mAh g^{-1} at a current density of 50 mA g^{-1} in the first cycle with a Coulombic efficiency of ≈99%. Zhang et al. utilized naturally derived dopamine (DA) to prepare a polydopamine (PDA) anode through oxidation, and achieved impressive capacities for Li^+ and Na^+ storage.[80] In another study, Wooley et al. introduced viologens and nitroxide radicals onto polypeptide backbones as the anode and cathode materials, respectively (Figure 7a).[81] Although the cytotoxicity tests demonstrated that the degradation products of these peptide-based electrode materials had low toxicity, the capacity of the assembled battery was only 37.8 mAh g^{-1}, indicating that further enhancement is needed. Although there is still a significant gap in capacity compared to traditional LIBs, when considering that biodegradable batteries primarily serve as power sources for wearable devices and medical sensors which do not require high capacity, they still hold broad prospects.[82]

Recently, Kang et al. developed fully degradable sodium-ion batteries (SIBs).[83] They used $Na_4Fe_3(PO_4)_2(P_2O_7)$ (NFP) as the cathode, a porous cellulose acetate (CA) mesh as the separator, thermoplastic protein-based carbon as the anode, CA and CMC as the binder, and a sodium perchlorate ($NaClO_4$) in a propylene carbonate (PC) solution as the electrolyte (Figure 7b). Each component of this battery could be decomposed into nontoxic compounds in nature (Figure 7c). Even when the used battery was buried in the soil for 120 days, plant growth was not affected (Figure 7d), well demonstrating its environmental safety. Moreover, their novel SIBs delivered a specific capacity of approximately 110 mAh g^{-1}, achieved a cycle retention of approximately 93% after 100 cycles at a current density of 20 mA g^{-1}, which is higher than that of the electrode using PVDF binders (≈89%).[83]

Despite notable advancements, biodegradable batteries often fall short of matching the capacity, energy density, and efficiency of traditional LIBs.[84] They are more suitable for specific applications with lower performance requirements, such as low-power portable electronics. However, producing biodegradable batteries presents difficulties, as current battery manufacturing processes are not compatible and require specialized equipment and new production methods. Additionally, while these batteries are designed to degrade after use, the environmental impact of their degradation products must be carefully evaluated to ensure they do not release toxic or harmful substances. Finally, establishing a robust infrastructure is also essential for the environmentally responsible disposal of biodegradable batteries.

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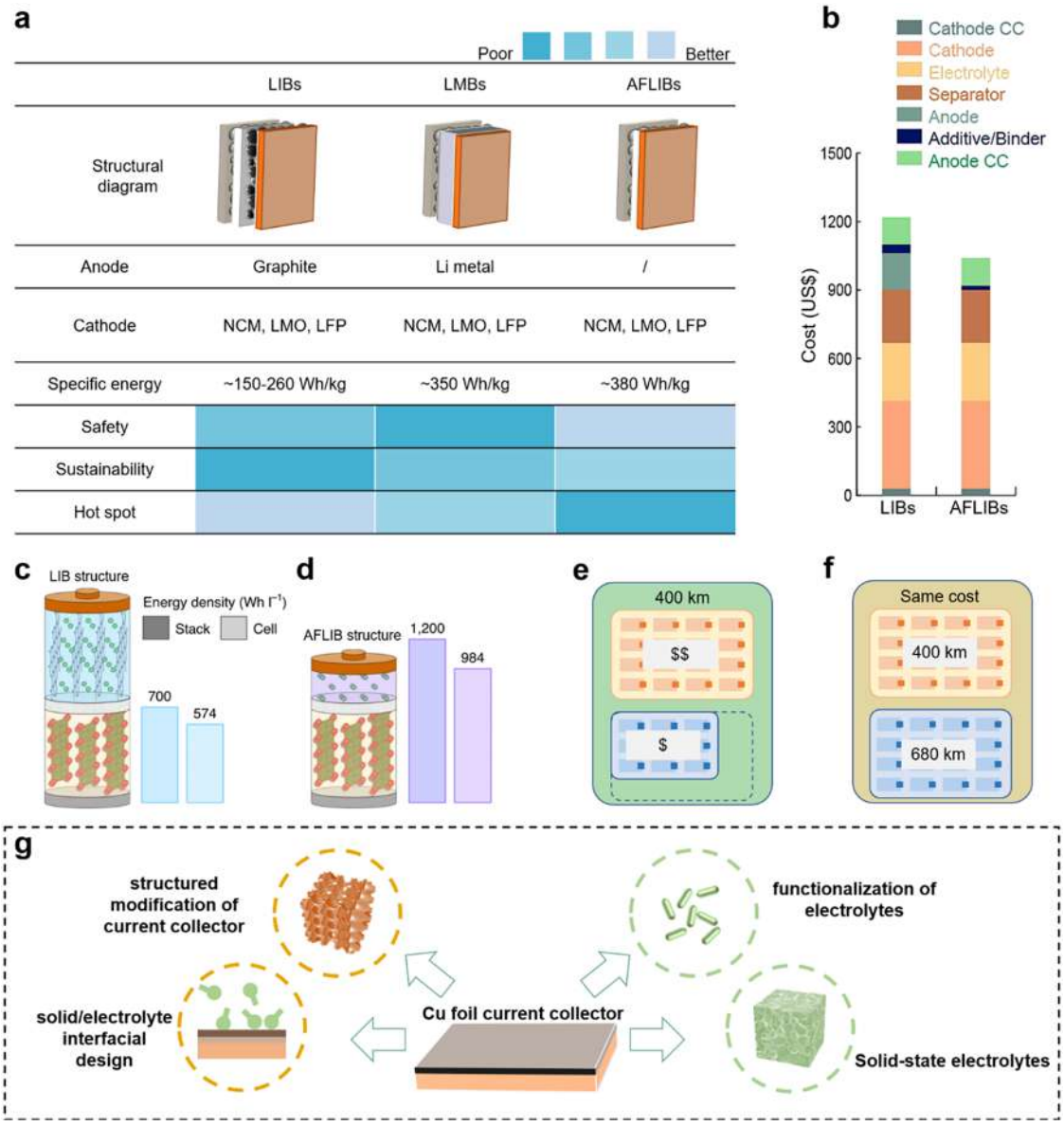

Figure 8. (a) Performance comparison for LIBs, LMBs and AFLIBs; (b) cost breakdown of different components for conventional LIBs and AFLIBs; (c, d) energy density comparison of LIBs and AFLIBs from cell and pack levels;[93] Adapted with permission from ref93. Copyright 2020 Springer Nature. (e) Required cell numbers of traditional LIBs and AFLIBs for the same driving distance;[93] (f) driving distances of the traditional LIBs and AFLIBs with same cell number;[93] (g) Potential solutions for addressing the Li dendrites issues of AFLIBs.

Anode-Free Batteries. Graphite is commonly used as the anode material in LIBs due to their high capacity and long cycle life. However, the mining and processing of graphite have significant environmental impacts, including habitat disruption, water pollution, and energy consumption.[85] Importantly, recycling used graphite anodes remains economically unfeasible, leading to their incineration during the recycling process, which generates environmental issues and hampers long-term sustainability.[1,86] To replace graphite, Li metal has been proposed as a next-generation anode material, which can offer a high specific energy density of approximately 350 Wh kg^{-1} (Figure 8a).[87] However, LIBs with Li metal anodes (LMBs) generally have a Coulombic efficiency of less than 99%, much lower than the 99.9% typically achieved with graphite anodes after initial cycles, leading to short cycle life. In addition, the high reactivity of Li metal requires stringent control over O_2, H_2O, and CO_2 during storage, transportation, and production, which substantially increases manufacturing costs.[51,88,89]

Recently, Dahn and co-workers introduced a new design for anode-free LIBs (AFLIBs), in which the anode material is entirely removed, leaving only the current collector.[90] During charging, Li^+ ions from the cathode are plated onto the current collector, while during discharge, the Li metal dissolves and the Li^+ ions are reinserted into the cathode.[51,91,92] Chen et al. compared the cost breakdown of AFLIBs with that of traditional LIBs (Figure 8b). The elimination of the graphite anode reduced the manufacturing cost of AFLIBs by 13%. The simplified battery structure and reduced weight improved the specific energy density to 380 Wh kg^{-1} and increased the volumetric energy density by around 71%. As a result, the driving range of electric vehicle could extend by an additional 280 km (Figure 8c,d).[93] Thus, fewer AFLIB battery packs are needed for

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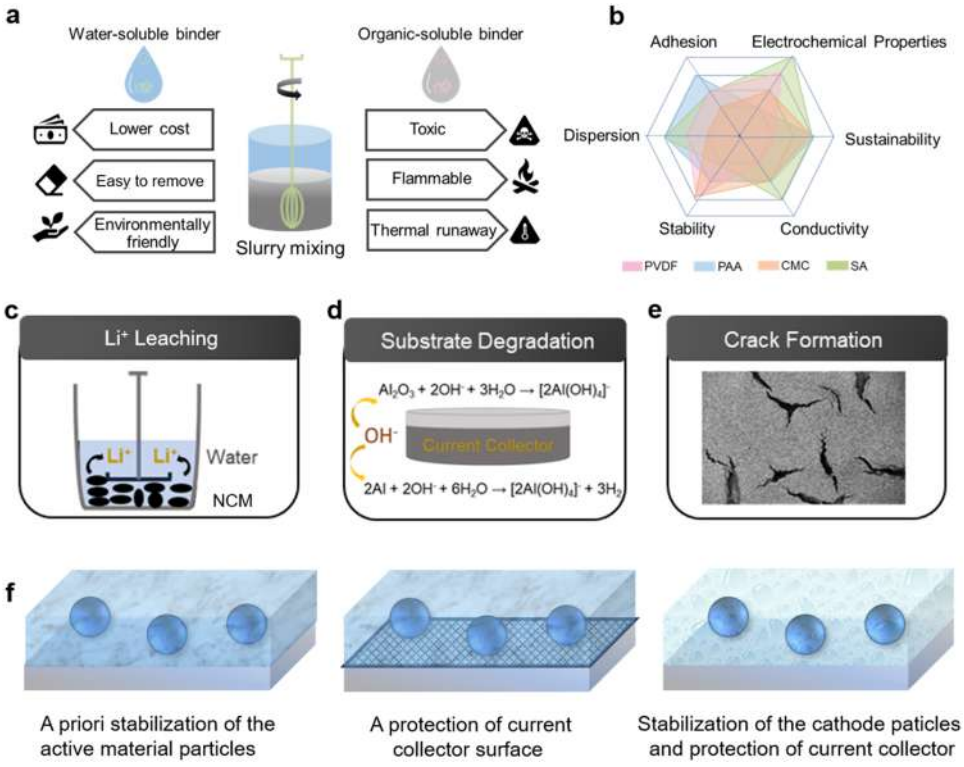

Figure 9. (a) Advantages of water-based binders and disadvantages of organic solvent-based binders; (b) comparison of the basic properties of PVDF and three commonly used water-based binders; (c–e) critical issues of using water-based binders for cathodes: Li^+ leaching, substrate corrosion, and crack formation; (f) potential strategies to address these issues: a priori stabilization of the active material particles, protection of current collector surface, stabilization of the cathode particles and protection of current collector.[117] Reproduced from ref 117. Available under a CC BY-NC license. Copyright Stefano Passerini.

the same range, providing considerable cost savings compared to conventional LIBs (Figure 8e).[93]

The removal of the graphite anode can streamline the recycling process for spent batteries. Although recycling for AFLIBs has not yet been proposed, their simplified structure eliminates the need for separating graphite from other components. It is important to note that a full discharge at a slower rate or with a lower voltage cutoff through electrochemical methods before recycling is crucial. This step helps dissolve the Li metal on the current collector and reintegrate it into the cathode, ensuring safe battery disassembly and components separation. Afterward, traditional recycling or regeneration methods can be applied to recover specific battery components effectively.

However, AFLIBs face comparable challenges similar to LMBs, including low Coulombic efficiency and dendrite formation.[94] Several strategies have been proposed to enhance Li^+ reversibility in AFLIBs, such as electrolytes functionalization, solid/electrolyte interfacial design, and current collector surface or structure modifications (Figure 8g). Dahn et al. recently reported a dual salt electrolyte composed of LiDFOB and $LiBF_4$,[90] enabling cells to retain 80% of their initial capacity after 90 cycles, a performance comparable to traditional LIBs. Abrha et al. introduced a Garnet-type $Li_7La_{2.75}Ca_{0.25}Zr_{1.75}Nb_{0.25}O_{12}$ (LLCZN) layer on the Cu current collector surface to prevent Li dendrite growth, and achieved an average Coulombic efficiency of 97.6% and a capacity retention of 58.66% after 30 cycles.[95] In addition, researchers have explored protective coatings or structural modifications of Cu current collectors to control the Li^+ flux and ensure smooth Li deposition. For instance, Chen et al. developed an AFLIB with a SiO_x-coated Cu current collector, which lasted for 60 cycles with 70% capacity retention, three times longer than the uncoated version.[96] Guo's team designed a 3D current collector with a submicron skeleton, enabling Li deposition within the 3D structure and preventing dendrite growth. Their AFLIB operated continuously for 600 h without short-circuiting, showing significantly improved safety and stability.[97]

However, significant challenges remain before the commercialization of AFLIBs, including technical limitations, safety concerns, and cost issues.[98] While AFLIBs outperform traditional LIBs regarding energy density, their limited cycling life poses a barrier to practical use.[99] Additionally, safety concerns resulted from Li dendrite growth and thermal runaway hinder widespread acceptance.[100] Strategies like functionalized electrolytes and SSEs have shown promise in extending AFLIB lifespan and improving safety, but these approaches tend to increase manufacturing costs. Therefore, further research is essential to develop advanced technologies and materials that enhance AFLIB performance while maintaining cost efficiency.

Water-Soluble Binders. Binders play a crucial role in maintaining the structural and mechanical integrity of electrodes in LIBs. They ensure cohesion among the active material particles and between the active materials and current collectors during electrochemical cycling. PVDF is a commonly used binder for cathode preparation, where it is typically dispersed in an organic solvent like *N*-methyl-2-pyrrolidone (NMP) along with conductive agents and active materials to create a homogeneous slurry. However, this approach raises safety and environmental concerns, and increases manufacturing costs.[101]

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Table 2. Electrochemical Performances of Various Aqueous Binder-Based Cathodes

cathode material	binder	initial discharge capacity[a] (mAh g^{-1})	rate capability, capacity[a] (mAh g^{-1})	cycle life, capacity[b] (mAh g^{-1})	refs
$LiFePO_4$	CMC	156.2 (C/5)	90 (5C)	152.2 (100)	He et al.[118]
$LiFePO_4$	hybrid humics/ CMC	153.6 (C/5)	105 (8C)	152.3 (100)	Yang et al.[119]
$LiFePO_4$	PTFE	150.3 (C/5)	88.6 (5C)	146.5 (100)	Gao et al.[120]
$LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$	Na-CMC	157.5 (C/2)	107.9 (5C)	90.1 (200)	Xu et al.[121]
$LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$	alginate	141.1 (C/2)	76.8 (5C)	89.2 (200)	Xu et al.[121]
$LiNi_{0.4}Co_{0.2}Mn_{0.4}O_2$	CMC	161.1 (C/10)	78.4 (10C)	91.6 (200)	Chen et al.[122]
$LiNi_{0.5}Mn_{1.5}O_4$	SA	118 (1C)	100 (5C)	86 (200)	Bigoni et al.[123]
$LiNi_{0.5}Mn_{1.5}O_4$	CCTS	135 (C/2)	95.8 (10C)	95.8 (100)	Zhong et al.[124]
$LiNi_{0.5}Mn_{1.5}O_4$	P(MVE−LMA)	126 (1C)	111.8 (10C)	92 (400)	Dong et al.[125]

[a]C-rate is given in parentheses. [b]Number of cycles is given in parentheses.

Furthermore, PVDF poses great challenges in spent LIBs recycling, as it is difficult to completely remove from the battery black mass, leading to low metal ion leaching efficiency. To enhance recycling efficiency, a pretreatment process involving low-temperature calcination is used to pyrolyze PVDF, though this process can release hazardous gases such as hydrogen fluoride (HF).

Recent research has focused on water-soluble alternatives to PVDF, which are less costly, more environmentally benign, and easier to remove during recycling (Figure 9a). These binders offer a significantly lower price range compared to PVDF ($2−$5/kg versus $12−$20/kg). In addition, water-soluble binders eliminate the dependence on toxic organic solvents, reducing environmental harm during electrode production. A LCA assessment demonstrated that replacing NMP with water could reduce CO_2 equivalent emissions by approximately 16%.[102,103] Furthermore, water-soluble binders can be easily washed away with water, simplifying the recycling process for LIBs.

Current choices for water-based binders include carboxymethyl cellulose (CMC), poly(acrylic acid) (PAA), and sodium alginate (SA).[104] As shown in Figure 9b, these binders demonstrate superior conductivity compared to traditional PVDF. Among them, CMC stands out as one of the primary binders used in water-based electrode manufacturing due to its high stability and is often used in combination with styrene−butadiene rubber (SBR) to enhance the binding strength and flexibility of electrodes.[105] PAA offers excellent dispersibility in water, tending to form a network structure that facilitates rapid transport of Li^+, which can improve the rate performance of batteries.[106−108] SA, derived from brown algae, possesses remarkable mechanical properties and minimal reactivity with the electrolytes. Additionally, each monomer unit in its polymer chain contains carboxyl functional groups, which contributes to its excellent water solubility.[109,110]

While the use of water-soluble binders for anode preparation has been commercialized, their application for cathode preparation is still under exploration. Zhang et al. conducted a comprehensive study on various water-soluble binders and their mixture for preparing LFP cathodes.[111] They found that electrodes made with SA and a combination of CMC and poly(tetrafluoroethylene) (PTFE) binders exhibited commendable performance. The rate capabilities (defined as the ratio of specific capacity at a 2C rate to that at a 0.1C rate) were measured to be 86.3% and 85.7% respectively, representing increases of 4.4% and 3.8% compared to electrodes using PVDF as a binder. After 50 cycles, the discharge specific capacities stabilized at 165 and 166 mAh g^{-1}, respectively. These impressive electrochemical performances are primarily attributed to the reduced polarization and improved electrochemical kinetic properties in the electrodes.[112] Notably, their results indicated that slurry viscosity plays a critical role in electrode performance, highlighting the necessity of including an appropriate amount of thickener, such as PTFE, SBR.

In contrast, the application of water-soluble binders for NCM electrode processing remains quite challenging. The highly reactive Li^+ in NCM lattice can be easily leached out by H^+ in aqueous slurry (Figure 9c), which not only leads to capacity loss of the NCM cathode but also damages the Al current collector due to increased slurry alkalinity (Figure 9d). Moreover, fabricating thick NCM electrodes with water-soluble binders is problematic, as cracks often develop due to capillary stress during water evaporation (Figure 9e).[113] Mukherjee et al. found that cracks appeared and propagated when the mass loading of NCM exceeded ∼15 mg cm^{-2}, resulting in delamination of the active materials during cycling.[113]

Several strategies have been proposed to mitigate Li^+ leaching during aqueous electrode processing (Figure 9f). For example, Matsumoto et al. created a water-resistant layer of TiO_x on the surface of $LiNi_aCo_bAl_{1-a-b}O_2$ ($a > 0.85$, NCA) particles, which effectively prevented Li^+ leaching during electrode preparation.[114] The resulting electrodes delivered a comparable rate performance to those made with PVDF binders. Furthermore, Al current collectors require protection, and applying a carbon coating can physically shield the Al foil from the aqueous slurry, enhancing both charge transfer kinetics and cycling performance.[115,116] Alternatively, combining these two strategies may provide a more effective approach to improve the stability of electrode stability. It is important to note that, although cathode electrodes prepared with water-soluble binders have achieved high electrochemical performances to date (Table 2), most studies have been limited to laboratory-scale button cells with low loading materials. Further investigation is needed to evaluate the electrochemical performance of full cells with higher-quality loaded electrodes and extended cycling tests.

CONCLUSIONS AND OUTLOOK

In conclusion, our thorough assessment of the environmental impact of conventional LIBs highlights the urgent need for sustainable advancements in battery technology. The development of metal-free electrodes, nonmetallic current collectors, the transition from LEs to SSEs, and anode-free batteries, along with the substitution of fluorine-based binders with water-soluble alternatives, are of great significance for reducing mineral

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dependencies and mitigating the environmental footprint of battery production in alignment with sustainable principles. Particularly noteworthy is the ambitious yet imperative vision of biodegradable batteries. These batteries, when fully realized, have the potential to revolutionize the energy storage landscape, effectively addressing concerns related to waste batteries accumulation and environmental pollution. Despite the substantial progress achieved in research and development, the widespread adoption of these advanced technologies still faces formidable challenges.

Performance. While the strategies outlined here can significantly enhance the sustainable development of LIBs, desirable performance remains a critical concern. For instance, metal-free electrodes often struggle with insufficient electrical conductivity, limiting their capacity and overall performance. Moreover, these electrodes may undergo structural transformations or degrade over multiple charge−discharge cycles, thereby diminishing battery longevity. Anode-free batteries, despite offering high gravimetric energy density, encounter critical issues related to their limited cycling life.

Scalability. Most of the leading advancements are demonstrated in laboratory-scale experiments. Transitioning these technologies to large-scale applications presents substantial challenges related to technological limitations and cost considerations. For instance, while SSEs can enhance battery safety and performance, their high production costs may pose significant barriers to widespread adoption. Although substituting metallic current collectors with carbon-based materials can effectively reduce the need for mineral mining, achieving large-scale production of electrodes using carbon-based current collectors while maintaining the same overall properties as traditional LIBs remains a technical hurdle. While water-soluble binders offer an environmentally friendly and cost-effective solution, the successful preparation of thick electrodes necessary for ensuring high energy density in batteries has yet to be realized. Addressing these challenges requires collaboration of between academic and industry efforts, focusing on the optimization of manufacturing processes, and developing new production techniques.

Compatibility. Compatibility, particularly concerning SSEs, poses a critical challenge. Insufficient alignment between SSEs and electrodes regarding ionic conductivity at the interface, thermal expansion coefficients, and mechanical stability has prevented all-solid-state batteries from reaching their projected potential. Overcoming these challenges necessitates a multifaceted approach involving precise material selection, interface engineering, and optimization of manufacturing processes. Researchers can focus on exploring diverse material combinations, interface modifications, and deposition techniques to enhance interfacial compatibility, thereby enabling the development of high-performance and robust solid-state batteries.

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Notes

The authors declare no competing financial interest.

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