

Review

Research Progress of Electrodes, Electrolytes and Interfaces in Solid-State Batteries

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Abstract: Solid-state batteries have become a critical growth area for next-generation energy storage systems, driven by their inherent benefits of superior operational safety, exceptional theoretical energy density, and extended cycle life. This work provides a systematic overview of recent progress in solid-state battery research, organized around three core components: electrode materials, electrolytes, and interphase engineering. In terms of electrode design, commercially dominant cathode systems primarily include lithium iron phosphate (LiFePO₄) and high-nickel nickel-cobalt-manganese (NCM) oxides, while anode development is heavily concentrated on lithium metal and carbon-based architectures. Each material class offers unique technical advantages but also suffers from intrinsic drawbacks, requiring tailored modification approaches to overcome their respective performance limitations. Electrolyte platforms are broadly categorized into three main families: polymer, oxide, and sulfide-based systems. For polymer electrolytes in particular, this review analyzes both free-standing electrolyte membranes and cathode-integrated coating layers, two configurations that exhibit distinct suitability for diverse application requirements. Interphase regulation represents the most significant barrier to the practical deployment of solid-state batteries. Longstanding challenges including poor solid-solid interfacial contact, elevated interfacial resistance, and undesired parasitic reactions severely degrade cell performance. These issues can be effectively alleviated through monolithic manufacturing techniques and targeted interphase modification strategies.

Keywords: electrodes; electrolytes; interfaces; solid-state batteries

1. Introduction

Lithium-ion batteries are ubiquitous in modern daily life [1]. Unfortunately, conventional liquid lithium-ion batteries suffer from a high risk of thermal runaway under high-temperature conditions, leading to prominent safety hazards. As a viable alternative, solid-state batteries (SSBs) use non-flammable solid electrolytes to replace liquid electrolytes, which fundamentally resolves the problems of electrolyte leakage and combustion. In addition, the robust mechanical properties of solid electrolytes can inhibit lithium dendrite propagation. Thanks to their high energy density and superior safety performance, SSBs have emerged as one of the most promising next-generation energy storage technologies. Currently, two major bottlenecks restrict the development of SSBs: unsatisfactory electrode-electrolyte interfacial stability and insufficient ionic conductivity of solid electrolytes. Breaking these limitations relies on high-conductivity lithium-ion materials, targeted interfacial modification strategies and advanced manufacturing techniques [2]. Unlike existing literature, this review centers on integrated solid-state battery systems. It systematically reviews the state-of-the-art research, technical challenges and optimization methods for electrodes, electrolytes and interfacial regulation—the three core components of SSBs, and elaborates



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on their underlying working mechanisms. This work also summarizes the prevailing challenges and future research directions in this field [3]. It can serve as a comprehensive reference for both basic research and industrial practice, and facilitate the commercialization of high-performance solid-state batteries. In addition to material performance deficits, SSBs are plagued by multiple practical issues such as interfacial chemo-mechanical instability, thermal safety risks and high manufacturing barriers, all of which impede their large-scale application [4]. Following the development roadmap, this review places emphasis on electrodes, solid electrolytes and interfacial engineering, and discusses related optimization strategies and emerging research trends.

2. Material Design—Types of Electrodes

Solid-state batteries (SSBs) replace flammable liquid electrolytes with solid alternatives, which fundamentally eliminates leakage and thermal runaway hazards. Benefiting from higher energy density, extended cycle life and broader operating temperature windows, SSBs show promising prospects for electric vehicles, portable electronics and large-scale energy storage. In SSB systems, electrode design must be tailored to match the ion transport behavior of solid electrolytes. Generally, composite cathodes are composed of active materials, conductive additives and binders: active materials are responsible for lithium ion storage and migration; conductive agents build continuous electron-ion pathways; binders preserve electrode structure and stabilize solid-solid interfaces. Every electrode type presents inevitable performance trade-offs [5]. Traditional electrodes represented by LFP and graphite have well-established manufacturing processes but deliver unsatisfactory energy density. In comparison, innovative electrodes and configurations such as sulfur cathodes, silicon-based anodes and anode-free designs enable high energy output, but they are troubled by drastic volume expansion, polysulfide shuttling, inhomogeneous lithium plating and dead lithium formation. These drawbacks become exacerbated under long cycling and harsh working conditions, posing critical challenges to practical applications. The overall performance of these electrodes is summarized in the Table 1. The following sections will conduct an in-depth discussion on the features, technical bottlenecks and modification approaches of LFP, NCM, lithium metal and graphite electrodes.

Table 1. Overview of mainstream electrode materials and novel configurations for solid-state batteries.

Electrode Type	Representative Material	Core Advantages	Main Drawbacks
LFP	LiFePO ₄	High thermal stability (decomposition at >700 °C), low cost, long cycle life (>3000 cycles).	Low electronic conductivity (10^{-9} – 10^{-10} S cm ⁻¹) and a relatively low operating voltage (~3.4 V vs. Li ⁺ /Li) [6].
Ni-rich NCM	NCM811, NCM90505	High theoretical capacity (>250 mAh g ⁻¹) and energy density (>1237.4 Wh kg ⁻¹).	Poor structural and thermal stability at high Ni content (decomposition at <200 °C) [7].
High-voltage spinel (LNMO)	LiNi _{0.5} Mn _{1.5} O ₄	Ultra-high working voltage (main plateau: 4.7 V vs. Li ⁺ /Li) [8].	Prone to electrolyte oxidative decomposition at high voltage; severe Mn dissolution and unstable CEI formation.
Sulfur cathode	Elemental sulfur	Ultra-high capacity (1675 mAh g ⁻¹), achievable energy density: 500~600 Wh kg ⁻¹ , low cost [9].	Low conductivity, polysulfide shuttling, ~80% volume expansion [10].
Lithium metal	Lithium foil	Ultra high specific capacity: 3860 mAh g ⁻¹ .	Dendrite growth, volume expansion [11].
Graphite	Graphite	Mature process, stable cycling.	Lithium plating and dendrite risk under fast charging; unstable SEI and particle damage [12].
Silicon-based anode	Silicon	Ultra-high theoretical capacity (~4200 mAh g ⁻¹), dendrite-free.	Huge volume expansion (>300%); low conductivity; unstable SEI and low ICE [13].
Anode-free design	Bare Cu/Al current collector	Greatly improved energy density; simplified process and high safety.	Uneven Li dendrites, large volume change, dead Li and low CE [14].

2.1. LiFePO₄ Cathode

LiFePO₄ is one of the most successful cathode materials, offering a theoretical specific capacity of 170 mAh g⁻¹, excellent thermal stability, and a high thermal decomposition temperature of 700–800 °C. It releases minimal gas during decomposition and presents a low risk of thermal runaway. The material exhibits stable electrochemical performance, particularly in terms of cycling performance, with a theoretical cycle number of more than 2000 cycles and practical applications reaching 3000–5000 cycles, making it well suited for long service life such as energy

storage and power tools. Wei X et al. [15] further optimized the volumetric capacity of LFP through a hot isostatic pressing (HIP) process. The influence of pressure on electrode structure and electrochemical performance can be directly observed through characterizations: As shown in Figure 1a, as the HIP pressure increases from 0 to 240 MPa, the density of the LFP cathode gradually rises from 2.2 g/cm³ to 2.62 g/cm³, while the porosity decreases from 40% to 23.5%, effectively reducing voids along the ion transport pathways. This structural densification directly translates into a significant improvement in volumetric capacity. Figure 1b presents the volumetric charge-discharge curves of HIP-treated LFP cathodes at 0.1 C, with the discharge volumetric capacity reaching 362 mAh/cm³, approximately twice that of conventional LFP cathodes without HIP treatment. For rate performance, which is a key bottleneck of LFP, Figure 1c shows the schematic of olivine-type crystal structure of LiMPO₄. Despite these advantages, LiFePO₄ has several intrinsic limitations, including low electronic conductivity on the order of 10⁻⁹–10⁻¹⁰ S cm⁻¹ and a relatively low redox potential. Additionally, the olivine-type structure of LiFePO₄ represents a typical one-dimensional tunnel structure. Its crystal lattice is composed of FeO₆ octahedra and PO₄ tetrahedra connected through shared vertices to form a three-dimensional framework, with Li⁺ occupying octahedral interstitial sites. This one-dimensional migration pathway fundamentally limits the Li⁺ transport rate [16], as illustrated in Figure 1d. Nevertheless, lithium iron phosphate has attracted attention as a cathode material. It combines a high theoretical capacity of 170 mAh g⁻¹, good thermal stability, and long-term cycling performance, with a reversible electrode potential of approximately 3.5 V, rendering it suitable for use with hybrid electrolytes. Moreover, LFP is composed of abundant iron resources and is environmentally benign. However, its poor electronic conductivity restricts electron transfer efficiency, leading to low rate performance. Under high charge-discharge rates, the capacity and overall performance of the battery are significantly compromised, thereby limiting its practical application. To overcome these challenges, researchers have explored various strategies. Debabrata Mohanty et al. [17] proposed the incorporation of lithium salts such as LiTFSI and conductive agents including Super P, KS-4. Their findings indicate that mixing with conductive agents effectively enhances the performance of LFP. The addition of LiTFSI not only reduces the overall battery resistance, mitigates impediments to internal Li⁺ transport, and lowers interfacial impedance but also enhances the discharge capacity and cycling stability of the battery. When the cathode contains 3% LiTFSI, the discharge capacity at 0.1 C reaches 168.9 mAh g⁻¹ at ambient temperature, close to the theoretical value. Even at a high charge-discharge rate of 0.5C, the battery maintains a high capacity of 161.8 mAh g⁻¹, with a capacity retention rate of 94%. From an overall development perspective, LFP has gradually become the preferred cathode for long-life, cost-oriented solid-state battery scenarios such as grid energy storage and commercial vehicles. Future research on LFP-based solid electrodes will continue to focus on structural densification, composite modification and compatible formulation design to further unleash its performance potential.

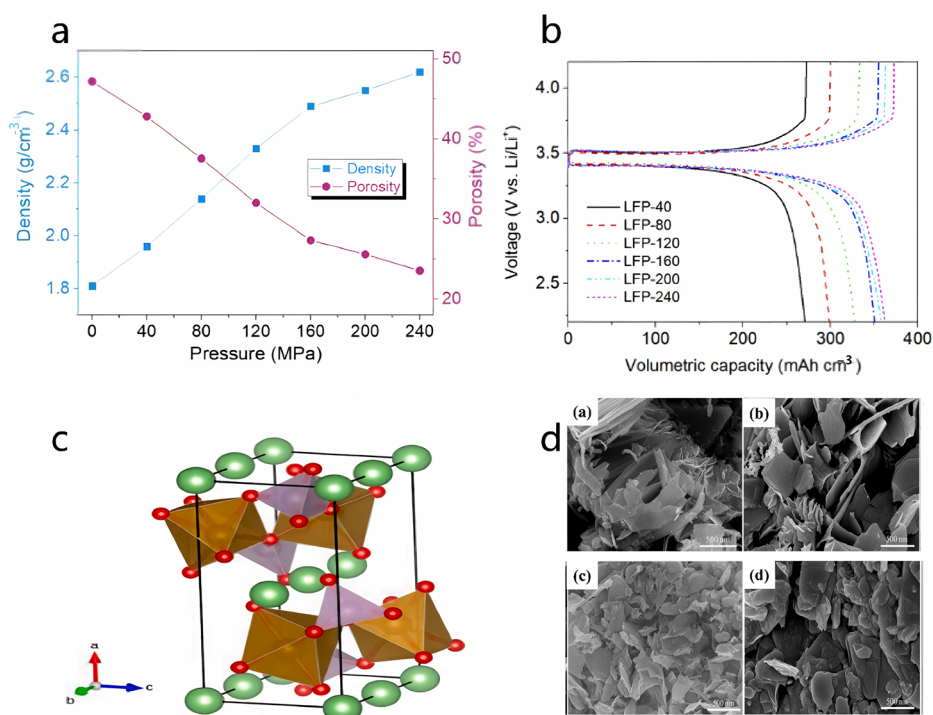


Figure 1. (a) Relationship between density/porosity of LFP cathodes and HIP pressure. (b) Volumetric charge-discharge curves of HIP-treated LFP cathodes at 0.1 C [14] Adapted with permission from Ref. [14]. Copyright

2024, Elsevier. (c) Schematic of olivine-type crystal structure of LiMPO_4 . (d) Morphology of $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ nanoplates with different nitrogen-doped carbon contents, as a typical polyanion phosphate material, it shares similar crystal structure and Li^+ transport mechanism with LFP [16]. Adapted with permission from Ref. [16]. Copyright 2024, Elsevier.

2.2. Nickel Cobalt Manganese Cathode

NCM electrodes are key cathode materials for lithium-ion batteries, composed of lithium and oxides of three transition metals: Ni, Co and Mn. Common types with different molar ratios include NCM111, NCM523, and NCM811. The specific capacity increases with the rise in nickel content [18], rising from approximately 140 mAh g^{-1} for NCM111 to 230 mAh g^{-1} for NCM90505, though this is accompanied by a decline in structural stability. The layered structure limits ion migration rate, therefore, its conductivity and cycling life must be improved through strategies such as nanonization, doping with Al^{3+} or Zr^{4+} , and surface coating with materials like $\text{Al}_2\text{O}_3/\text{LiNbO}_3$. Compared with LiFePO_4 , LiMn_2O_4 , and other cathode materials, NCM achieves a synergistic effect in which Ni boosts capacity, Co stabilizes the layered crystal structure, and Mn enhances structural stability. This combination enables both higher stability and capacity, making NCM widely used in portable electronic devices and electric vehicles, as exemplified by the Tesla 4680 battery, which achieves an energy density of 250–300 Wh/kg using high-nickel NCM. Nevertheless, NCM faces challenges including high-temperature capacity fading, poor thermal stability with a decomposition temperature of approximately 200°C , and interfacial side reactions. Future research should focus on single crystallization, compatibility with solid electrolyte, and sustainable production technologies [19]. The preparation of NCM also presents technical difficulties. The CSTR coprecipitation method commonly used in industry requires precise control over multiple parameters and generates substantial waste. The solid-state conversion method, on the other hand, often suffers from uneven mixing due to insufficient diffusion of metal ions, which compromises electrochemical performance. To address these issues, Won-Gwang Lim, Kyuchul Shin et al. [20] adopted a solid-state conversion method using low-melting-point metal oleate precursors, enabling homogeneous mixing of metal ions at the molecular level. The resulting NCM-OA exhibits improved electrochemical performance, including higher discharge capacity and Coulombic efficiency (CE). Overall, high-nickel NCM will remain the core research direction for high-energy solid-state cathodes, while compatibility with various solid electrolytes is a long-term research priority.

2.3. Lithium Metal Anode

As the core anode material for solid-state batteries, lithium metal has a body-centered cubic crystal structure, an ultrahigh theoretical specific capacity of 3860 mAh g^{-1} , and an extremely low electrochemical potential of -3.04 V (vs. standard hydrogen electrode, SHE). When combined with solid electrolytes such as oxides, sulfides, or halides, it enables battery energy density exceeding 400 Wh/kg. Leveraging the mechanical strength and thermal stability of solid electrolytes, lithium metal fundamentally eliminates the risks of electrolyte leakage and thermal runaway associated with liquid batteries, while also exhibiting compatibility with high-voltage cathodes to further enhance energy output. Despite these advantages, several challenges hinder its practical application. Lithium dendrites tend to propagate along grain boundaries or defects within solid electrolytes during charge-discharge processes, potentially causing short circuits. Chemical incompatibility with certain solid electrolytes, such as sulfide and NASICON-type materials, triggers interfacial side reactions that lead to the formation of a high-impedance solid electrolyte interphase layer. Additionally, volume expansion during cycling can result in interfacial delamination and poor contact. To address these challenges, modification strategies have been developed primarily around interface optimization, electrolyte enhancement, and structural design. Interfacial chemistry can be regulated by applying protective layers such as LiPON or LiF, or by introducing alloy layers such as Li-Mg or Li-In, thereby improving compatibility and suppressing dendrite growth [21]. Solid electrolytes can be modified through approaches such as doping with Ga^{3+} to stabilize the cubic phase of LLZO [22], preparing composite electrolytes based on ceramic-polymer composite systems [23], or achieving densification via hot pressing sintering and the use of sintering aids to enhance ionic conductivity and mechanical strength [24]. Structural strategies include designing 3D porous electrolyte frameworks and applying external pressure to expand the electrode-electrolyte contact area, thereby mitigating the effects of volume expansion. Lithium alloying further contributes to suppressing dendrite growth and preventing structural degradation [25], ultimately improving the cycling stability and safety of solid-state batteries. Looking ahead, the synergistic optimization of interface, electrolyte and electrode structure will be the mainstream direction for lithium metal anodes, and its large-scale application depends on the comprehensive solution of dendrite and interfacial failure problems.

2.4. Graphite Anode

As a classic core anode material for lithium-ion batteries, graphite features a hexagonal layered crystal structure, a practical theoretical specific capacity of 372 mAh g^{-1} , and a low electrochemical potential of approximately 0.1 V (vs. Li/Li^+). When combined with organic liquid electrolytes or solid electrolytes, it provides stable and compatible energy output for commercial lithium-ion batteries, making it the most mature anode material for industrial application to date [26]. Graphite enables the reversible intercalation and deintercalation of Li^+ through its layered structure, exhibiting excellent electrochemical stability and a flat voltage plateau during charge-discharge processes. It demonstrates strong compatibility with various cathode materials, including NCM, LFP, and NCA. In addition, graphite benefits from abundant raw material reserves and a well-established preparation and modification process system, striking a favorable balance between electrochemical performance and industrial viability. However, its application faces several challenges. The repeated intercalation and deintercalation of lithium during long-term charge-discharge processes induce electrochemical stress, leading to microcracks and even structural fracture of graphite particles. Electrolyte decomposition results in the formation of a thick, porous, and high-impedance SEI layer on the graphite surface, contributing to capacity fading and reduced CE. Additionally, excessively fast Li^+ intercalation rate at high rates triggers metallic lithium deposition. Beyond performance issues, the recycling and regeneration of spent graphite face practical challenges, including high energy consumption and elevated temperature in pyrometallurgical processes, severe pollution from hydrometallurgical methods, and the formation of loose amorphous carbon coatings during low-temperature modification [27]. To address these challenges, modification strategies have focused on structural repair, surface modification, and optimization of recycling and regeneration processes: Carbon coating modification, using materials such as turbostratic or amorphous carbon, can fill microcracks in graphite particles and facilitate the construction of artificial SEI layers, thereby inhibiting electrolyte side reactions and metallic lithium deposition [28]; The effectiveness of such targeted repair strategies has been verified through experimental characterizations [26]: Figure 2a presents ICP analysis results of metal impurity content in spent graphite (SG), purified graphite (PG-1), and PG-2. The data show that the contents of Fe, Cu, Mn, Co, Ni, and other impurities in PG-1 are reduced to $\leq 5 \text{ ppm}$, significantly lower than those in SG, effectively eliminating interfacial side reactions caused by impurities. In terms of cycling stability, Figure 2b compares the cycling performance of SG, PG-CR-8 wt% (graphite repaired with 8 wt% carbon coating), and commercial graphite (CG) at 1 C . After 450 cycles, PG-CR-8 wt% maintains a capacity of 355 mAh g^{-1} with a capacity retention rate of over 98%, substantially outperforming SG and approaching the cycling stability of CG, confirming the effectiveness of carbon coating in repairing structural defects in graphite. The high initial CE of regenerated graphite (RG) is attributed to the reconstruction of the SEI layer and the retention of active lithium. Figure 2c shows the initial CE of CG, damaged graphite (DG), and RG at different SOC (10%, 20%, 30%). At 20% SOC, the initial CE of RG reaches 104.7%, significantly higher than that of CG at 87.3%, clearly demonstrating the effectiveness of this repair scheme in improving interface stability and reducing active lithium loss. Additional strategies include converting carbon precursors such as sucrose into dense turbostratic carbon coatings via a low-temperature magnesium-catalyzed process to reduce the graphitization activation energy and improve the ionic and electronic conductivity of graphite [29]. For spent graphite, a combination of acid washing for impurity removal and carbon coating modification enables the repair of structural defects and the regeneration of electrochemical performance. Meanwhile, optimizing the recycling process to avoid high-temperature smelting and excessive use of strong acids and alkalis not only enhances the performance of RG but also balances the economic and environmental sustainability of the recycling process. Graphite anode research mainly includes surface optimization and spent graphite regeneration. Carbon modification and impurity removal can restore the performance of recycled graphite. As a low-cost material, graphite will continue to be applied in medium- and low-energy solid-state batteries, and green recycling is an important research topic.

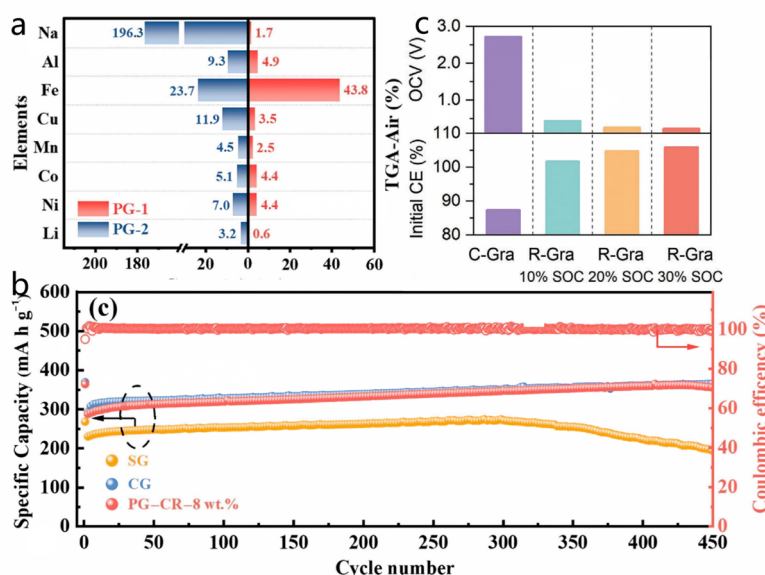


Figure 2. (a) ICP analysis of metal impurity content in SG, PG-1 and PG-2. (b) Cycling performance comparison of SG at 1 C, PG-CR-8 wt% and CG at 1 C [26] Adapted with permission from Ref. [26]. 2024, Elsevier. (c) Initial CE comparison of CG, DG and RG [28] Reprinted/adapted with permission from Ref. [28]. 2024, Wiley-VCH.

3. Electrolyte Engineering

Solid electrolytes are the core components of solid-state lithium-ion batteries. To date, three main types have been extensively investigated: oxide electrolytes, sulfide electrolytes, and polymer electrolytes, each offering distinct advantages and disadvantages. Garnet-type oxide solid electrolytes exhibit high Li⁺ conductivity at room temperature and excellent thermal stability, enabling rapid Li⁺ transport and outstanding safety. However, they suffer from poor interfacial compatibility with electrodes and involve complex preparation processes [30]. Sulfide solid electrolytes also possess high Li⁺ conductivity but are hindered by high costs and insufficient interfacial stability [31,32]. Table 2 compares the primary advantages and limitations of polymer, oxide-based and sulfide-based solid electrolytes. Since this work centers on polymer electrolytes and integrated electrode-electrolyte structures, subsequent content will focus exclusively on polymer electrolytes. Polymer electrolytes offer notable advantages in flexibility and excellent chemical compatibility with electrode materials, yet they are limited by low Li⁺ conductivity and a narrow electrochemical stability window [33]. Based on the bonding mode with cathodes, polymer electrolytes can be further divided into two core forms: freestanding electrolyte membranes exist as independent sheets and are sandwiched between cathodes and anodes through methods such as stacking or hot pressing. This approach features mature processes and strong universality, though interfacial contact still requires further optimization; cathode-coated electrolyte membranes are directly deposited onto the cathode surface as a thin coating, forming a cathode-electrolyte integrated structure that offers low interfacial impedance and good adaptability to high-loading cathodes, while imposing stringent requirements on coating uniformity. The following sections will elaborate on polymer solid electrolytes and these two membrane configurations in detail.

Table 2. Comparison of typical advantages and limitations of three types of solid-state electrolytes.

Component	Representative Material Systems	Core Advantages	Main Drawbacks
Oxide-based	Garnet-type Li ₇ La ₃ Zr ₂ O ₁₂ (LLZO)	Excellent chemical stability [34].	Require ultra-high sintering temperature (>1000 °C) [35], Severe mechanical brittleness.
Sulfide-based	Li ₁₀ GeP ₂ S ₁₂ (LGPS), Li ₆ PS ₅ Cl (LPSCI)	Ultra-high ionic conductivity (>10 ⁻³ S cm ⁻¹) [36].	Thermodynamically unstable at the interfaces of Li metal anodes and high-voltage cathodes [37].
Polymer electrolytes	PEO-LiTFSI	Superior interfacial contact with electrodes, Good flexibility.	Low ionic conductivity at room and medium temperatures, Narrow electrochemical window (4.0–4.2 V vs. Li ⁺ /Li) [38].

3.1. Polymer Electrolytes

As core components of lithium-ion batteries, polymer electrolytes (PEs) consist of lithium salts dispersed in a polymer matrix and are generally classified into three categories: gel polymer electrolytes (GPEs), solid polymer electrolytes (SPEs), and inorganic filler-reinforced PEs. Their structural characteristics are primarily reflected in their ion conduction mechanisms and key physicochemical properties. Owing to their easy processability and high mechanical strength, they represent a major research focus in the field of solid electrolytes [39]. Among these, polyethylene oxide (PEO) has emerged as the most widely studied polymer solid electrolyte due to its advantages such as low reactivity with electrodes, high flexibility, and excellent redox stability. As a high molecular polymer, PEO is synthesized via ring-opening polymerization of ethylene oxide monomers, with a repeating unit of $-\text{CH}_2\text{CH}_2\text{O}-$. Multiple repeating units are connected by covalent bonds to form a long-chain molecular structure, and the ether bonds in PEO serve as important carriers for Li^+ migration [40]. However, PEO exhibits drawbacks such as poor mechanical properties and high crystallinity, which significantly restrict segment motion and consequently impair ion conduction efficiency [41]. Ion conduction in polymer electrolytes relies on the segment motion of polymer chains and the dissociation of lithium salts, with key performance parameters including ionic conductivity (target $\geq 10^{-4} \text{ S cm}^{-1}$), Li^+ transference number, mechanical modulus ($\geq 6.8 \text{ GPa}$), and an electrochemical stable window of 4–5 V [42–44]. They offer remarkable performance advantages: intrinsic safety eliminates the risk of leakage and combustion associated with liquid electrolytes; they can function as both electrolytes and separators, reducing the total mass of the battery and thereby improving energy density; and their excellent mechanical flexibility makes them suitable for the design of flexible and irregular batteries [45]. Nevertheless, polymer electrolytes face multiple challenges. At the interface level, the porous structure of cathodes results in insufficient electrolyte wetting, the volume change of electrodes during charge-discharge processes creates interfacial gaps, and anodes suffer from high impedance issues induced by lithium dendrite growth and space charge layers (SCL) [46]. In terms of performance, traditional polymer electrolytes, such as those based on PEO, exhibit low ionic conductivity at room-temperature, a trade-off between mechanical strength and ion conduction, and insufficient thermal stability at high temperatures. To address these challenges, modification strategies focus on interface optimization and electrolyte engineering, leading to numerous key technological breakthroughs. Three main approaches are employed to improve the mechanical properties of PEO [47]. The first involves the introduction of plasticizers, which increase the amorphous phase content of polymers, thereby enhancing segmental motion and promoting the dissociation of lithium salts. The second is blending with other polymers, where PEO is combined with low-crystallinity polymers to improve the mechanical properties [48]. The third is the formation of composite electrolytes by incorporating oxide electrolytes, leveraging the characteristics of inorganic fillers to enhance both overall mechanical strength and ion conduction efficiency. In recent studies, Jabbari et al. [49] prepared a GPE with a room-temperature ionic conductivity of $1.5 \times 10^{-3} \text{ S cm}^{-1}$ by immobilizing a liquid electrolyte within a methacrylic acid-modified PEO cross-linked network. A Li/LFP battery with this GPE maintained a capacity retention rate of 70% and a CE of approximately 99% after 1000 cycles at 2C. Guan et al. [50] constructed a composite solid electrolyte with a room-temperature ionic conductivity of $3.3 \times 10^{-4} \text{ S cm}^{-1}$ and a Li^+ transference number of 0.82 by introducing imide-linked COF (NKCOF-29) into a fluorinated polyimide (FPI) matrix via homologous imide bonds interactions, effectively eliminating phase separation. Xiao et al. [51] introduced the dielectric material BaTiO_3 (BTO) into in-situ polymerized CPEs, regulating the Li^+ coordination structure through the interaction between BTO and the polymer ether groups to reduce the interfacial impedance drop. The Li||NCM811 battery achieved 1800 cycles at room temperature under 4.6 V and 1300 cycles under 4.7 V. Qu et al. [52] designed a fluoropolymer-based plastic crystal electrolyte (FPCE) via interface engineering, SN molecules using the cross-linked network of HFBA and PETEA to induce the formation of a LiF-rich hybrid SEI. A Li|Cu battery with this FPCE exhibited a CE of 98.25% after 300 cycles at 0.5 mA cm^{-2} . These modification strategies address the application bottlenecks of polymer electrolytes from multiple perspectives, including structural design, interface regulation, and component optimization. In particular, the modification and composite strategies applied to PEO have further expanded its potential for application in high-energy-density lithium-ion batteries.

After elaborating on the basic composition, ion conduction mechanism and general limitations of polymer electrolytes (PEs), we further classify them according to their forming structures and assembly modes with electrodes. Two typical configurations are widely adopted in current research: freestanding solid electrolyte membranes (independent films) and cathode-electrolyte integrated membranes (integrated films). To clearly distinguish their structural characteristics, preparation technologies and electrochemical performances, the key differences between the two configurations are briefly summarized in Table 3.

As shown in Table 3, the freestanding solid electrolyte membrane is the most widely used configuration due to its versatile performance and mature manufacturing process. Its detailed properties and research progress are discussed in the following section.

Table 3. Key characteristics comparison of two typical polymer electrolyte configurations.

Configuration	Core Advantages	Main Drawbacks
Freestanding Solid Electrolyte Membranes	Controllable film thickness [53] easy production Satisfactory mechanical strength [54]	Unavoidable solid-solid contact gaps, which increase interfacial resistance [55].
Cathode-Electrolyte Integrated Membranes	Ultra-tight electrode-electrolyte contact [56] Efficient lithium-ion transport [57] Streamlined fabrication	Strict requirements for slurry rheology and coating uniformity, which hinder scalable manufacturing [58].

3.2. Freestanding Solid Electrolyte Membranes

Polymer freestanding electrolyte membranes are independent sheets and are sandwiched between cathodes and anodes via stacking or hot pressing. Their core structure consists of lithium salts (e.g., LiTFSI, LiFSI) loaded in a continuous polymer matrix (e.g., PEO, PVDF-HFP, PAN). Some formulations incorporate inorganic fillers (e.g., inert SiO₂, active LLZTO) to construct a synergistic system [59]. Structurally, these membranes rely on the coordination of Li⁺ with polar groups such as ether oxygen and cyano groups, combined with segment motion of polymer chains, to facilitate ion conduction. In PEO-based membranes, the repeating unit is -CH₂CH₂O-, with ether bonds serving as the key carriers for Li⁺ migration. In composite systems, inorganic fillers can inhibit polymer crystallization and establish continuous ion transport channels [60]. Such membranes must possess the mechanical strength and ion transport capacity required for self-supporting formability, with key performance parameters including room-temperature ionic conductivity (target $\geq 10^{-4}$ S cm⁻¹), Li⁺ transference number and an electrochemical stability window of 4–5 V [61]. Polymer electrolyte membranes offer significant performance advantages: excellent flexibility and processability, the ability to serve as both electrolytes and separators to reduce the total battery mass for higher energy density, strong universality that ensures compatibility with different cathode systems such as NCM and LFP without requiring cathode-specific formulation optimization, mature large-scale production processes, and facile and precise control over thickness and dimensions [62]. However, they also face multiple challenges: the restricted segment motion of polymers leads to low ionic conductivity (e.g., pure PEO-based membranes are usually below 10^{-5} S cm⁻¹) at room temperature [60]; physical stacking contact with electrodes readily induces the formation of a space charge layer (SCL), leading to increased interfacial impedance; a trade-off exists between mechanical strength and ion conduction, necessitating modification to pure polymer membranes to resist lithium dendrites [63,64]; adequate thermal stability at high temperatures and a narrow electrochemical stability window in some systems further limit compatibility with high-voltage cathodes [65]. To solve these problems, several modification strategies have been proposed, including: introducing lithium salts with large anions such as LiTFSI and LiFSI to promote dissociation and increase the concentration of free Li⁺ via a weak coordination effect [66]; adding inert inorganic fillers (e.g., SiO₂, Al₂O₃) or active inorganic fillers (e.g., LLZTO, LATP) to inhibit polymer crystallization, enlarge the amorphous regions, and create additional ion transport pathways [67]; improving mechanical strength and interfacial stability through cross-linking polymerization (e.g., PEO cross-linked network) or polymer blending (e.g., PEO-PVDF-HFP); and accelerating ion diffusion by employing the Lewis acid coordination strategy (e.g., the interaction between Mg²⁺/Al³⁺ and EO chains) to weaken the Li⁺-EO interaction [68]. Relevant research progress has been made in this area. Huang et al. [69] developed a self-healing composite solid electrolyte based on reversibly interlocked polymer networks (RILNs), achieving a room-temperature ionic conductivity of 1.06×10^{-3} S cm⁻¹ when compounding with LGPS fillers. Liu et al. [70] prepared a Co-MOF filled PEO-based composite electrolyte, in which Co²⁺ promoted the dissociation of LiTFSI, enabling stable cycling for more than 120 cycles at room temperature. Duan et al. designed a PEO-based composite membrane modified with a PVDF-HFP interlayer, which significantly reduced the interfacial impedance and allowed stable Li⁺ stripping/plating for more than 1000 h.

3.3. Cathode-Electrolyte Integrated Membranes

The separate preparation of electrolyte membranes often leads to unstable electrode-electrolyte interfaces, and the stability of solid-solid interfaces consistently falls short of that of solid-liquid interfaces [71]. To address this issue, researchers coat the electrode surface with a liquid electrolyte to allow it to penetrate the electrodes and form a thin film, thereby constructing an electrode-electrolyte integrated structure before assembling with the counter electrode. This approach effectively improves interfacial contact. The core feature of such integrated

polymer electrolytes is the formation of a tightly coupled structure comprising a conductive network-active particles-electrolyte [72]. In a typical configuration, polymers such as PEO and polyacrylonitrile (PAN) serve as the matrix to build a three-dimensional porous framework, enabling the electrolyte slurry or precursor to uniformly fill the pores. This provides directional transport channels for Li^+ and reduces unnecessary transport pathways. In the integrated structure, no obvious gaps exist at the electrolyte-electrode interface, and the components are uniformly dispersed, thereby optimizing ion transport efficiency [73].

Integrated polymer electrolytes exhibit remarkable performance advantages. First, they offer significantly improved interfacial stability. Given the relatively small volume change of cathode materials during charge-discharge processes, integration with solid electrolytes helps preserve interfacial integrity, mitigate contact degradation caused by volume change, and thus extend the battery cycling life. Second, they demonstrate excellent electrochemical performance, with outstanding room-temperature ionic conductivity and Li^+ transference number, alongside an electrochemical stability window as wide as 0–5 V, rendering them compatible with most high-voltage active materials. Third, they combine good flexibility with structural robustness. Some integrated pouch batteries maintain a capacity retention rate of more than 95% after 1000 bends, meeting the requirements of flexible electronic devices [74]. Fourth, they exhibit excellent rate performance. A high discharge specific capacity of 117 mAh g^{-1} can be maintained at a 3C, with a CE close to 99.8% and a charge transfer resistance only 1/15 of that of traditional structures [75,76]. Substantial research progress has been made in this area. Yang et al. [77] developed vertically aligned composite polymer electrodes, which significantly improved ion conduction efficiency of high-areal-capacity solid-state batteries by rationally designing fast ion conduction pathways. Chen et al. [74] designed a cathode-supported electrolyte membrane framework that enhanced the electrode-electrolyte interfacial contact through structural optimization, thereby further improving overall battery performance. The structural advantages of this cathode-supported integrated framework are clearly illustrated in the following figures. Figure 3a compares the architecture of liquid LIB, conventional SSLIB and cathode-supported SSLIB. In the integrated structure, the electrolyte is directly cast onto the cathode surface, forming a gap-free interface with enhanced contact and improved safety, thereby addressing the poor wettability typically encountered in traditional solid-state batteries. Figure 3b presents a cross-sectional SEM image of the cathode-supported solid electrolyte membrane, clearly showing tight bonding among the cathode layer, electrolyte layer, and current collector, with no obvious delamination or gaps. The structural integrity directly contributes to low interfacial impedance. Deng et al. [72] developed carbon nanotube network integrated electrodes, achieving conformal coating of active particles and electrolytes, which substantially reduces interfacial impedance and significantly improves battery cycling stability. Figure 3c presents a schematic of the layer-by-layer assembly process for the CNT-enveloped integrated electrode.

Despite the prominent advantages of integrated polymer electrolytes, their practical application still faces multiple challenges. First, the challenge of anode interface compatibility. The volume change of silicon-based and other anode materials during charge-discharge processes can reach up to 300%. Such substantial expansion and contraction generate stress at the solid electrolyte-anode interface, damaging the interfacial stability and leading to hindered ion transport [78]. Second, inherent limitations of electrolytes themselves persist. Some polymer electrolytes (e.g., PEGDMA) are prone to brittle fracture and insufficient mechanical strength after polymerization. Moreover, reliance on initiators like azobisisobutyronitrile (AIBN) may introduce organic residues that affect the structural stability of solid-state batteries [79]. Third, strong temperature dependence remains a concern. The ionic conductivity and mechanical strength of PEO-based frameworks are highly temperature-sensitive, and the restricted segment motion at low temperatures leads to a sharp decline in electrochemical performance [80]. Fourth, there is the risk of interfacial side reactions. Multiphase diffusion between electrodes and electrolytes can trigger chemical reactions, resulting in the formation of a high-impedance interfacial layer and hindering charge transport [73]. Collectively, these issues severely restrict the large-scale application of integrated polymer electrolytes, highlighting an urgent need to overcome these bottlenecks through targeted modification strategies.

To address the above challenges, researchers have developed a series of efficient modification strategies focusing on multi-dimensional optimization for core issues such as anode volume change, insufficient electrolyte performance, and interfacial side reactions. Notable progress has been achieved through the following approaches: constructing a high-toughness LiF-PEO electrolyte framework to buffer volume expansion and inhibit interfacial delamination by leveraging its excellent flexibility and ductility; meanwhile, doping PEO with LiF to reduce crystallinity, increase the amorphous phase content, and enhance lithium salt dissociation capacity for faster Li^+ transport [73]; introducing a CNTs network as a dual framework for conduction and support, which not only strengthens the structural stability of electrodes but also optimizes the electron transport pathways and reduces interfacial impedance [80]; modifying the cathode surface with a lithium niobium oxide coating to inhibit transition metal dissolution and electrolyte oxidative decomposition, thereby improving compatibility with PEO-based

electrolytes [81]; designing a bilayer system composed of a PEO/LiDFOB cathode contact layer and a PEO/LiTFSI anode layer to balance the chemical stability of different interfaces [82]; and optimizing the in-situ polymerization process to reduce the dosage of organic solvents and catalyst residues, thereby improving the purity and mechanical strength of electrolytes. Outstanding research achievements have been reported in this area. The lithium niobium oxide coating modification strategy developed by Liang et al. [81] effectively stabilized the interface between Ni-based cathodes and PEO-based electrolytes, with the battery maintaining a capacity retention rate of 94% after 250 cycles. Figure 3d shows the HAADF-STEM images of ALD-LNO coated NCM811 after 50 cycles. The PEO-based composite electrolyte prepared by Xue et al. [73] exhibited improved temperature adaptability through LiF doping and polymer blending, achieving a room-temperature ionic conductivity increased to $0.43 \times 10^{-4} \text{ S cm}^{-1}$. Huo et al. [80] developed a cationic metal-organic framework filled anion-immobilized polymer electrolyte capable of effectively inhibiting lithium dendrite growth, enabling stable cycling of lithium metal batteries for 600 h. Yu et al. [83] designed a composite polymer-ceramic electrolyte that simultaneously improved mechanical strength and ionic conductivity by virtue of the synergistic effect of ceramic fillers and polymer matrix, providing a feasible path for the practical application of the integrated configurations.

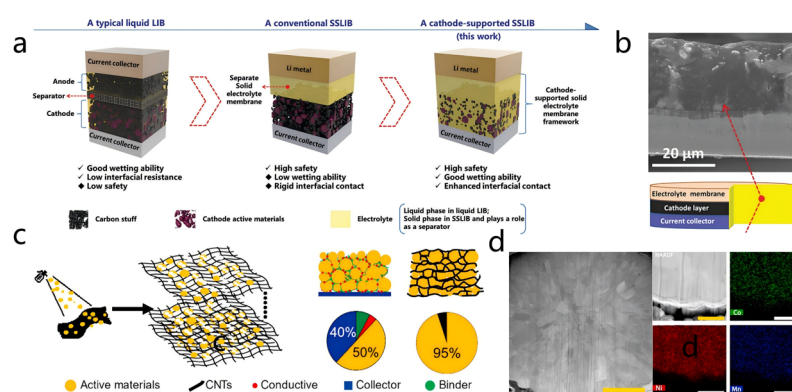


Figure 3. (a) Schematic comparison of liquid LIB, conventional SSLIB and cathode-supported integrated SSLIB. (b) Cross-sectional SEM image of cathode-supported solid electrolyte membrane [74] Adapted with permission from Ref. [74]. Copyright 2019, Royal Society of Chemistry. (c) Schematic of layer-by-layer assembly for CNTs-enveloped integrated electrode [72] Reprinted/adapted with permission from Ref. [72]. Copyright 2021, Elsevier. (d) HAADF-STEM images of ALD-LNO coated NCM811 after 50 cycles [81] Reprinted/adapted with permission from Ref. [81]. Copyright 2020, Elsevier.

Cathode-electrolyte integration represents a core strategy for addressing the challenges of poor interfacial contact and high impedance in solid-state batteries. Its essence lies in achieving molecular-level fusion between the electrolyte and cathodes through process innovation, thereby eliminating the interfacial defects inherent to traditional discrete structures. In light of the growing trend toward solvent-free dry film technology and existing research results, the current mainstream integration methods are mainly powder spraying (including electrostatic spraying), binder fibrillation, and cathode-supported tape casting. Each method features distinct principles, performance characteristics, and application scenarios, collectively advancing the optimization of interfacial performance in solid-state batteries [84].

As one of the core solvent-free integration technologies, the powder spraying method uses solvent-free mixture powders as raw materials. The mixture comprising cathode active materials, electrolytes, conductive agents, and binders is deposited onto a current collector via electrostatic or mechanical spraying, followed by hot pressing and curing to form an integrated structure. This method offers remarkable advantages: no solvent pollution, compatibility with solvent-sensitive electrolytes such as sulfides to avoid active material hydrolysis and electrolyte performance attenuation; excellent uniformity of coating thickness, enabling the preparation of thick electrodes of 100–200 μm to improve energy density; and compatibility with roll-to-roll production, suitable for large-scale mass production. Li Y et al. [85] elaborated on the powder spraying method in detail in their review, and its process principle and parameter optimization effects are as follows: Figure 4a presents a schematic diagram of the powder spray process for integrated electrode fabrication, which realizes solvent-free integrated molding through two steps of powder application and hot rolling fixation, and is compatible with roll-to-roll large-scale production. However, this method also has obvious shortcomings: the problem of powder overflow and waste leads to limited powder utilization efficiency; stringent requirements are imposed on equipment precision and powder characteristics, necessitating precise control of parameters such as spraying voltage (about 15 kV) and

powder feeding pressure (≥ 15 psi); additionally, powder particle size and dielectric constant are significantly affected by environmental humidity, requiring pre-treatment and drying [86]. In relevant research, Ludwig et al. [87] prepared LiCoO_2 cathode-electrolyte integrated electrodes via electrostatic spraying combined with a hot pressing process. Compared with the wet process, these electrodes exhibited higher bonding strength with the current collector, avoided the coverage of active sites by binders, and exhibited significantly improved electrochemical performance. Liu et al. [88] adopted a “binder priming and powder spraying” strategy, first spraying a PVDF binder layer on the current collector and then depositing the active material-electrolyte mixed powders. This approach increased the bonding strength of graphite electrodes by 166% and improved the capacity retention after 100 cycles from 24.2% to 92.4%. The resulting electrolyte layer of the prepared sulfide solid-state battery is less than 50 μm , with high active material loading and excellent rate performance [89].

The binder fibrillation method involves uniformly mixing cathode active materials, electrolytes, and conductive agents with fibrillatable binders such as PTFE. High shear forces are applied using equipment like jet mills and screw extruders, causing the binder to form a nanoscale fiber network that entangles and immobilizes the various components. This mixture is then formed into an integrated self-supporting membrane through hot pressing [90]. Figure 4b,c schematic illustrate the binder fibrillation process for integrated film formation and the one-step electrospinning/electrostatic spraying for integrated composite electrolyte, respectively. The core advantages of this method are: extremely low binder dosage (<1 wt%), which avoids insulating binders from obstructing ion transport; the fiber network can construct a three-dimensional electron-ion dual transport channel, balancing mechanical strength and interfacial continuity; and the solvent-free process can protect electrolyte activity, especially suitable for sensitive systems such as sulfides and halides. However, this method is extremely sensitive to process parameters.

Insufficient shear force leads to inadequate binder fibrillation, while excessive shear force may damage the electrode active material coating. In addition, binder compatibility is limited, and traditional materials such as PTFE are prone to side reactions at low voltages. Large-scale production requires precise control over the uniformity of the fiber network and involves high equipment investment costs. Several research cases demonstrate the potential of this approach. Hippauf et al. [91] realized PTFE fibrillation via manual grinding to prepare an integrated cathode based on a sulfide electrolyte with only 0.3 wt% binder; the assembled pouch-type solid-state battery maintained a capacity retention rate of 93.6% after 100 cycles. Figure 4d presents a schematic diagram of the DF process, which forms a self-supporting membrane using only 0.1–1 wt% PTFE binder through a solvent-free process of mixing, shear-induced film formation, and cold pressing composite, thereby avoiding obstruction of ion transport by insulating binders. Anna et al. regulated the molecular weight of PIB binder. In the resulting PIB-LPSCI composite integrated membrane, the high-molecular-weight binder formed a denser fiber entanglement network, significantly optimizing the membrane self-supporting property and critical current density (CCD). Wang et al. [92] prepared a large-size LLZTO integrated membrane of 8×6 cm^2 with a thickness of only 15–20 μm via a rolling-coordinated fibrillation process. A pouch cell matched with NCM811 cathode achieved an energy density of 280 Wh/kg. Gao L et al. [90] developed an integrated composite electrolyte prepared by a one-step electrospinning/electrostatic spraying process, which exhibits excellent interface stability and high-voltage adaptability. Cycling performance of Li/PMIA-PEO-e/Li symmetric cells at different current densities showed stable voltage for 800 h without fluctuation, verifying the long-term interface compatibility between the integrated electrolyte and Li metal anode.

The cathode-supported tape casting method adopts a double-layer tape casting process: a cathode tape is first prepared via tape casting, after which the electrolyte slurry is directly cast onto the surface of the uncured cathode. Following co-curing, a cathode-supported integrated electrolyte membrane (CSSEM) framework is formed [93]. This method offers several notable advantages: strong interfacial wettability and adhesion, realizing seamless bonding between the electrolyte and cathode via in-situ casting, greatly reducing interfacial impedance; a simple process that is compatible with existing tape casting equipment without requiring additional complex devices, making it suitable for low-cost, large-scale production; and the ability to stack multiple layers to assemble multi-cell battery packs, showing great potential for large-scale application [94]. However, this method also has limitations: the rheological properties of the electrolyte slurry must be well matched with those of the cathode tape, otherwise interlayer delamination is prone to occur; in addition, the method is currently mainly compatible with PEO-based composite electrolytes, and its tape casting compatibility with inorganic electrolytes is still further optimized. Xinzhì Chen et al. [93] prepared a CSSEM framework using PEO-based composite electrolyte as the raw material via air tape casting; compared with traditional solid-state batteries, the battery employing this structure exhibited a 10–15% increase in discharge capacity at different temperatures and rates, a reduction in interfacial resistance of over 40%, and maintains excellent cycling stability after stacking 2–3 cells [94].

In summary, the powder spraying method focuses on uniformity and compatibility with mass production, the binder fibrillation method has the advantages of low binder dosage and flexible adaptability, and the cathode-supported tape casting method is well suited for low-cost, traditional electrolyte systems. Each method is adapted to different application scenarios. Future research should focus on optimizing process parameters, developing specialized binders, and localizing equipment manufacturing to further overcome cost and performance bottlenecks, thereby promoting the large-scale application of cathode-electrolyte integration technology [95].

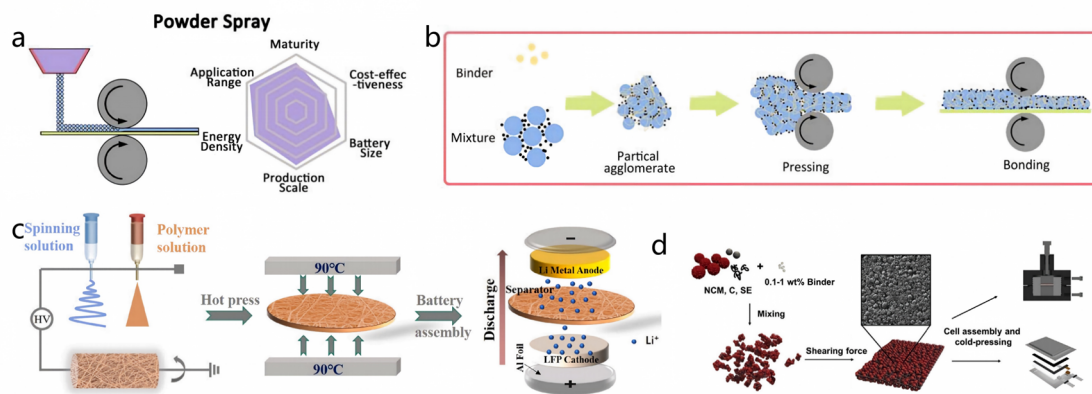


Figure 4. (a) Schematic procedure of powder spray for integrated electrode fabrication [85]. Reprinted/adapted with permission from Ref. [85]. Copyright 2022, Elsevier. (b) Schematic of binder fibrillation process for integrated film formation. (c) Schematic of one-step electrospinning/electrostatic spraying for integrated composite electrolyte [90]. Reprinted/adapted with permission from Ref. [90]. Copyright 2021, Elsevier. (d) Schematic of solvent-free dry-film (DF) process for integrated cathode fabrication [91]. Reprinted/adapted with permission from Ref. [91]. Copyright 2019, Elsevier.

4. Interfacial Regulation

Interfacial challenges constitute the primary bottleneck for the commercialization of solid-state batteries. These issues originate from intrinsic drawbacks of solid-solid contact and progressive structural degradation of electrodes upon cycling, and are essentially categorized as coupled chemo-mechanical failures. From a mechanical perspective, electrode volume fluctuation and rigid contact of solid electrolytes readily give rise to interfacial voids and delamination during long-term cycling. Meanwhile, chemical degradation including parasitic side reactions, electrolyte decomposition and element interdiffusion continuously deteriorates interfacial stability, eventually causing increased impedance and capacity fading. For cathode systems, the conventional discrete electrode-electrolyte structure forms numerous gaps between solid electrolytes and active materials, blocking lithium ion transport and inducing unwanted interfacial side reactions. In terms of anodes, lithium metal tends to form penetrating dendrites, and silicon anodes experience a volume expansion of up to 300%. Both phenomena lead to severe interfacial delamination and stress buildup, along with a drastic increase in interfacial resistance [84]. In addition, reactions between electrolytes and impurities on electrode surfaces generate low-conductivity products and deplete active lithium, which further accelerates interfacial failure. Restricted solid-solid contact and the formation of space charge layers also hinder efficient lithium ion migration.

To address these challenges, researchers have developed multidimensional regulation strategies: the construction of inorganic or alloy-based interfacial protective layers to inhibit dendrite growth and side reactions [96]; optimization of electrode structure to buffer interfacial damage caused by volume expansion; improvement of the mechanical strength and uniformity of ion conduction in electrolyte through doping and composite modification [97]; introduction of interfacial buffer layers to enhance wettability and reduce interfacial impedance [98]; and the use of first-principles calculations combined with artificial intelligence (AI) to screen compatible materials for precise interfacial compatibility optimization [99]. The effectiveness of these strategies has been fully verified through experimental characterizations and theoretical simulations (Figure 5). Specifically, Sb-modified LLZO interfaces enable uniform lithium deposition and avoid dendrite formation [96] (Figure 5a); the synergistic combination of mechanical strength and ionic conductivity in electrolyte significantly inhibits the growth kinetics of lithium dendrites [97] (Figure 5b); the LLZTO/Li-Nafion dual-phase interface establishes stable ion transport channels, preserving electrode surface after prolonged cycling [98] (Figure 5c); and efficient screening of SEI materials with both high shear modulus and high ionic conductivity can be achieved through lateral transfer learning, providing new material candidates for interfacial regulation [99] (Figure 5d).

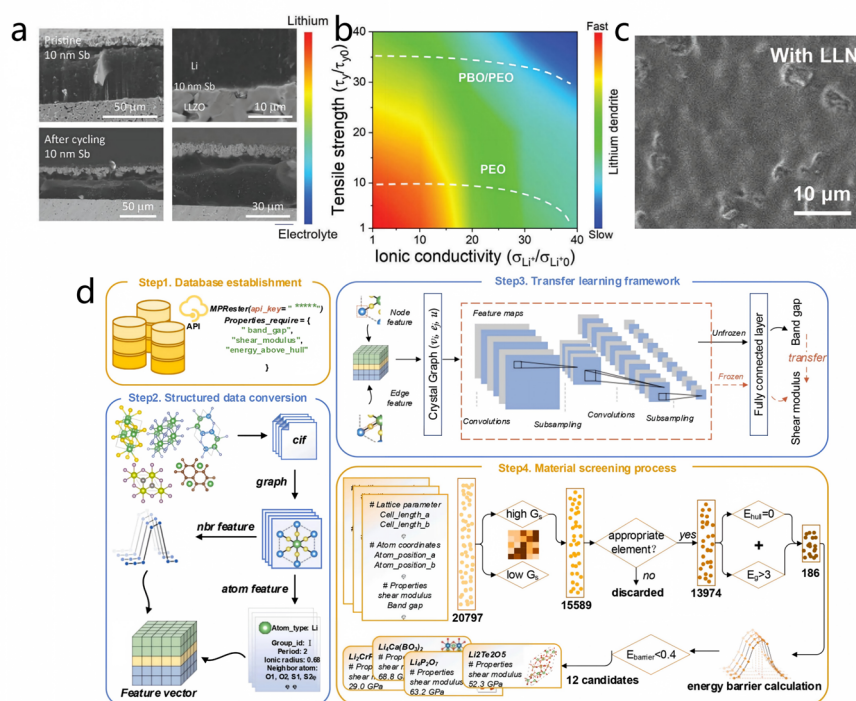


Figure 5. (a) SEM image of Li anode after cycling with Sb-modified LLZO interface [96]. Reprinted/adapted with permission from Ref. [96]. Copyright 2021, Wiley-VCH. (b) Simulation diagram of Li dendrite growth regulated by electrolyte mechanical strength and ionic conductivity [97]. Reprinted/adapted with permission from Ref. [97]. Copyright 2024, Wiley-VCH. (c) SEM image of Cu foil surface after cycling with LLZTO/Li-Nafion dual-phase interface protection [98]. Reprinted/adapted with permission from Ref. [98]. Copyright 2019, Wiley-VCH. (d) Workflow diagram of high-performance SEI material screening via lateral transfer learning [99]. Reprinted/adapted with permission from Ref. [99]. Copyright 2025, Elsevier.

To systematically solve interfacial issues, current research focuses on two core directions. The first is the integration design of the cathode-electrolyte interface, which achieves molecular-level fusion between the electrolyte and cathodes through techniques such as powder spraying and electrospinning. This approach enables the construction of gap-free interfaces, reducing impedance and inhibiting side reactions. The second is the precise regulation of the anode interfaces, targeting the inherent defects of lithium metal and silicon-based anodes, by employing strategies such as protective layer construction and structural optimization to mitigate volume effects and dendrite growth, and ensure long-term interfacial stability. Working synergistically from both cathode and anode sides, these two directions provide key solutions for improving the interfacial performance of solid-state batteries.

4.1. Cathode-Electrolyte Interfacial Regulation

The key challenges facing the current cathode-electrolyte interface exhibit system dependence. When combined with polymer electrolytes, the interface is prone to electrolyte decomposition under high voltages, such as the redox incompatibility between LCO and PEO. In contact with oxide electrolytes (e.g., LLZO, LATP), interdiffusion of elements such as Co, La, and Zr occurs during high-temperature sintering or cycling, leading to the formation of high-impedance intermediate phases. When matched with sulfide electrolytes, space charge layer effects and interfacial degradation become prominent, along with side reactions that generate low-conductivity products (e.g., Li_2S , Li_3P); moreover, poor solid-solid contact generally results in high interfacial impedance [52]. Currently, the cathode-electrolyte interface remains the core bottleneck restricting the practical application of solid-state batteries. First, high interfacial impedance caused by poor solid-solid contact runs through all systems. Due to differences in rigidity, polymer electrolytes (e.g., PEO) struggle to form continuous contact with cathodes (LCO, NCM). The physical rigidity of oxide electrolytes (LLZO, LATP) combined with the irregular morphology of cathode particles exacerbate interfacial gaps. Although sulfide electrolytes are relatively soft, mismatched surface energy with oxide cathodes still leads to contact defects. These gaps directly hinder Li^+ transport and amplify polarization [100,101]. Second, element interdiffusion and interfacial phase transformation are prominent. Studies have shown that Co, La, and Zr diffuse across the LCO and LLZO interface during high-temperature sintering or cycling, forming high-impedance amorphous layers and impurity phases such as LaCoO_3 . When NCM

is in contact with LATP, Li^+ deintercalates from NCM and enriches in LATP at high temperatures, inducing grain boundary phase transformation in NCM particles [102]. Third, frequent side reactions arise from electrochemical incompatibility. For example, when high-nickel NCM is compounded with PEO, active oxygen on the NCM surface readily causes oxidative decomposition of the ether bonds in PEO, and the released fluorine radicals further exacerbate transition metal dissolution [103]. Sulfide electrolytes (e.g., LGPS, LPSCI) are prone to cyclic decomposition within the operating voltage window of NCM and LCO, forming low-conductivity products such as Li_2S and Li_3P . Meanwhile, the space charge layer effect caused by differences in Li chemical potential further hinders charge transfer [104]. Fourth, dynamic instability during cycling poses a critical issue. The volume expansion and contraction of cathode materials upon cycling damage interfacial contact, while uneven growth or shedding of the SEI/CEI layers leads to a continuous increase in impedance [105]. For example, the CEI layer formed at the NCM and sulfide electrolytes interface lacks transition metal compounds, making long-term stable passivation difficult to achieve [106].

To address these challenges, current mainstream solutions focus on interfacial regulation and structural optimization. For instance, Li Q et al. [100] constructed a PTFEMA interfacial layer on the surface of NCM811 via in-situ solvent-free bulk electropolymerization. The $-\text{CF}_3$ groups in PTFEMA improve the high-voltage stability of PEO, enabling the electrolyte to achieve an ionic conductivity of $1.02 \times 10^{-4} \text{ S} \cdot \text{cm}^{-1}$, while the battery exhibits tight interfacial contact and significantly improved cycling stability. Shen et al. [101] modified NCM811 with a ZIF-8@ionic liquid composite coating and added ZIF-8 nanofillers to the PEO electrolyte. At 0.2 C and 60 °C, the discharge capacity reaches approximately 150 mAh g^{-1} , effectively inhibiting PEO decomposition and transition metal dissolution. Guo Q et al. [107] designed a PGII chelating copolymer interlayer to modify NCA, which utilizes the chelation between IDA groups and metal ions to reduce the interfacial impedance of the PVDF/LAGP system and achieve rapid Li^+ transport. Liu F et al. [108] prepared a gradient trilayer solid electrolyte (GTSSE), which enhances the interfacial compatibility between NCM811 and the electrolyte through the synergistic effect of PVDF-HFP and PEGDME while constructing continuous Li^+ transport channels. The resulting battery maintains a discharge capacity of 107.1 mAh g^{-1} after 1000 cycles. Chandra et al. [109] found that Co replaces the B-site in LLTO at 300 °C, inducing element mixing and significantly reduces the interfacial impedance.

4.2. Anode-Electrolyte Interfacial Regulation

Currently, the anode-electrolyte interface in solid-state batteries still faces four key challenges. First, there is the risk of short circuits caused by lithium dendrite growth along grain boundaries or defects, which is closely related to uneven interfacial ion transport and local stress accumulation [110]. Second, the volume expansion of silicon-based anodes during lithiation reaches up to 300%, and the dissolution and deposition of lithium metal anodes during cycling are accompanied by volume fluctuations, leading to interfacial delamination and a sharp increase in impedance [111]. Third, mismatched stress-strain behavior and electronic structure further exacerbate interfacial degradation. For example, electrolytes such as LLTO and LATP undergo reduction reactions upon contact with lithium metal, resulting in crack formation and increased grain boundary resistance [112]. Fourth, interfacial side reactions generate low-conductivity products, consuming active lithium and damaging interfacial contact; in some systems, gas evolution also occurs, leading to battery swelling [113]. To address these challenges, multi-dimensional innovative regulation strategies have been developed. Currently, the state of the art has achieved a capacity retention rate of over 80% for silicon-based anode full batteries after 600 cycles, stable cycling of lithium metal symmetric batteries for over 1500 h, interfacial impedance reduced to below 5 $\Omega \cdot \text{cm}^2$ in some systems, and a critical current density increased to 1.5 mA $\cdot \text{cm}^{-2}$.

Relevant teams have achieved significant breakthroughs through differentiated technical approaches. Huang et al. [100] revealed the interfacial reaction mechanism between silicon and sulfide electrolytes using first-principles calculations, finding that Si and P interact to form irreversible products such as SiP_2 and SiP , thereby providing theoretical support for the design of interfacial buffer layers. Xu et al. [114] developed a novel $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ through a Ge atom substitution strategy, increasing the Schottky barrier of the LGPS–Li interface to 0.13 eV and reducing the electron transfer region, thereby improving interfacial stability without compromising Li^+ conductivity. Sun et al. [115] prepared an inorganic-organic composite coating at the interface between silicon anodes and sulfide electrolytes using molecular layer deposition technology, significantly inhibiting interfacial side reactions and enabling the battery to maintain a capacity retention rate of 90% after 200 cycles. Ci et al. [116] introduced a LiAlO_2 coating with high ionic conductivity and mechanical strength on the silicon surface, which not only promotes Li^+ interfacial transport but also alleviates silicon volume expansion, effectively protecting the structural integrity of the electrode-electrolyte interface. Shin et al. [117] coated $\text{Li}_6\text{PS}_5\text{Cl}$ electrolyte onto the surface of two-dimensional conductive graphene-like carbon (GLC@LPSCI), inhibiting interfacial side reactions

and promoting uniform ion migration, leading to a significant improvement in battery cycling stability. Collectively, these studies have advanced the field from multiple dimensions including theoretical design, material innovation and process optimization, promoting continuous breakthroughs in the interfacial performance of anode-electrolyte interface.

5. Conclusions

Solid-state batteries represent a revolutionary energy storage technology that completely replaces liquid electrolytes with solid-state electrolytes, fundamentally eliminating safety risks such as electrolyte leakage and thermal runaway. Their core structure comprises cathodes, solid electrolytes, anodes, and current collectors working synergistically. Cathodes typically consist of active materials like LFP or high-nickel NCM, conductive agents, and solid electrolytes to enable continuous electron-ion transport [118]. Anodes center on lithium metal, offering ultrahigh capacity, and carbon-based graphite, which facilitates early commercialization. With advantages in safety, energy density exceeding 400 Wh/kg theoretically, and long cycle life, solid-state batteries are recognized as a key direction for next-generation energy storage, with broad applications in electric vehicles, energy storage stations, and consumer electronics [119].

Despite remarkable research progress, solid-state batteries still face four major technical hurdles. First, most solid electrolytes suffer from low ionic conductivity at room temperature. Second, poor solid-solid contact inevitably leads to rising interfacial impedance and severe electrochemical side reactions. Third, electrode materials encounter structural degradation during long-term cycling. Fourth, the large-scale manufacturing processes remain complicated and difficult to scale up [120]. Different application scenarios impose distinct requirements. Electric vehicles demand high energy density and stable interfaces, energy storage prioritizes cost-effectiveness and rate performance, while consumer electronics focus on miniaturization and flexibility [121]. Notably, Semi-solid-state batteries are now defined as hybrid solid-liquid batteries and serve as a vital transitional solution prior to the commercialization of solid-state batteries. Driven by continuous advances in material innovation, interface optimization and process upgrading, solid-state batteries deliver superior safety performance, higher energy density and longer cycle life [122]. From laboratory research to industrial technology implementation, the industry keeps promoting the development of solid-state batteries toward large-scale commercialization.

Author Contributions

Y.W.: conceptualization, literature retrieval and curation, writing—original draft preparation; J.Q.: literature screening, comparative data sorting; S.W.: visualization, table drawing; C.H.: reference collation, partial writing—reviewing; L.Z.: conceptualization, supervision, reviewing and editing; M.Z.: manuscript revision, funding acquisition, conceptualization, validation of research logic. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement

No new data were created or analyzed in this study.

Conflicts of Interest

The authors declare no conflict of interest.

Use of AI and AI-Assisted Technologies

No AI tools were utilized for this paper.

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